Dynamical Mean-Field Theory of Resonating Valence Bond Antiferromagnets

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We propose a theory of the spin dynamics of frustrated quantum antiferromagnets, which is based on an effective action for a plaquette embedded in a self-consistent bath. This approach, supplemented by a low-energy projection, is applied to the kagome antiferromagnet. We find that a spin-liquid regime extends to very low energy, in which local correlation functions have a slow decay in time, well described by a power law behaviour and $\omega/T$ scaling of the response function: $\chi''(\omega) \propto \omega^{-\alpha} F(\omega/T)$.

The possibility of frustrated quantum antiferromagnets (QAF) having a resonating valence bond (RVB) ground state, that is, a superposition of states where all spins are paired into singlets, was suggested many years ago [1], such a ground state has neither long-range spin order (e.g., Neel order) nor spin-Peierls order (an ordered arrangement of these singlet pairs). Recently, convincing evidence has been given that some frustrated two-dimensional QAF indeed have RVB physics [2,3]. In particular, numerical studies [3] of the spin-half Heisenberg QAF on the kagome lattice reveal that the model has no long-range order and displays a very small gap to magnetic (triplet) excitations (estimated to be of order $J/20$). Moreover, this gap is filled with an exponential number of singlet excitations, suggesting a possible continuum in the thermodynamic limit, and a large low temperature entropy, in agreement with the RVB picture. We also note, though this is seldom emphasized, that the results of [3] suggest a correspondingly large number of triplet states immediately above the gap. This may be expected, since in any valence bond component of an RVB state, one can replace any singlet pair by a triplet pair and continue to have an eigenstate of $S$. Resonance between such states would lower the excitation energy to much below $J$.

These considerations suggest that, in a temperature range above the triplet gap, the spin correlations have a rapid decay in space, but a slow decay in time due to the large density of triplet excited states. Equivalently, the dynamical susceptibility $\chi(\vec{q}, \omega)$ would not display a narrow peak around a specific ordering wave-vector, but $\chi''(\vec{q}, \omega)$ would have weight at low frequency, characteristic of a spin liquid. Indeed, recent inelastic neutron scattering studies [4] on the $S = 3/2$ kagome slab compound SCGO reveal such behaviour above the freezing temperature ($T \geq 4K$). The only relevant energy scale in this spin-liquid regime is apparently set by the temperature itself, with $\chi''_{\text{loc}} \equiv \sum_{\vec{q}} \chi''(\vec{q}, \omega)$ obeying a scaling behaviour $\chi''_{\text{loc}} \sim \omega^{-\alpha} F(\omega/T)$ with $\alpha \simeq 0.4$, corresponding to a slow decay in time of the local dynamical correlations $\langle S(x,0)S(x,t) \rangle \sim 1/t^{1-\alpha} \sim 1/t^{0.6}$. We observe that the uniform ($\vec{q} = \vec{0}$) susceptibility in this regime is well fitted by $\chi \propto T^{-0.4}$ too.

In this article, we introduce a novel theoretical approach to the spin dynamics in the spin-liquid regime of frustrated QAF, which focuses on short-range spin correlations only. This approach draws inspiration from the dynamical mean-field theory (DMFT) of itinerant fermion models [5], and some of its extensions [5-10]. Our method is fairly general but is applied here to the concrete case of the spin-half kagome QAF. The determination of on-site and nearest-neighbor dynamical spin correlations is mapped onto the solution of a model of quantum spins on a triangular plaquette coupled to a self-consistent bath. (A single-site approach is not adequate, since the essential physics of singlet formation involves at least nearest-neighbor sites). Using a projection onto the low-energy sector of this model, we demonstrate that, in a temperature range extending down to $T \ll J$, a slow (power-law) decay of temporal correlations is found.

![FIG. 1. The kagome lattice, viewed as an up-pointing cluster (bold) embedded in a network of similar clusters, which is approximated in the DMFT approach by a self-consistent bath.](image-url)

We view the kagome lattice as a triangular superlattice of up-pointing triangular plaquettes (Fig. 4). Sites are labeled by an index $a = 1, 2, 3$ within a plaquette and by a triangular superlattice index $I$ numbering the plaquette. We denote by $\chi_{ab}(\vec{q}, \tau - \tau')$ the Fourier transform of the dynamical spin correlation function $\frac{1}{3} \langle \vec{S}_{a,I}(\tau) \cdot \vec{S}_{b,I'}(\tau') \rangle$ with respect to the plaquette coordinates $I, I'$ ($\vec{q}$ is a vec-
tor of the supercell Brillouin zone). Our approach relies on approximating the correlation function by

$$\chi(\vec{q}, i\nu_n)^{-1}_{ab} = J_{ab}(\vec{q}) + \mathcal{M}_{ab}(i\nu_n).$$ \hfill (1)

Here, $\nu_n \equiv 2n\pi/\beta$ is a bosonic Matsubara frequency, and all quantities are matrices in the internal plaquette indices $(a, b)$. $J_{ab}(\vec{q})$ is the supercell Fourier transform of the exchange couplings. $\mathcal{M}_{ab}$ is a measure of how much the correlation function differs from that of a Gaussian model, for which $\chi = J^{-1}$, and hence plays the role of a spin “self-energy” matrix for the QAF. Obviously, the key assumption made in (1) is that the $\vec{q}$-dependence of this self-energy can be neglected. This approximation, which is likely to be reasonable when spatial correlations are short-ranged, is analogous to the assumption of a momentum-independent self energy made in the DMFT of correlated fermion models. Here however, the DMFT concept is extended on two accounts: the local ansatz is made on the spin correlation function rather than on a single-fermion quantity, and a plaquette rather than a single site is considered. Extensions of DMFT to clusters and to (spin or charge) response functions have been considered separately before in different contexts. Combining them is a unique aspect of our approach, which is necessary to capture the dynamical aspects of inter-site singlet formation at the heart of spin-liquid behaviour.

In order to calculate the dynamical self-energy matrix $\mathcal{M}_{ab}(i\nu_n)$, an effective action involving only the spins of a single triangular plaquette is introduced, which reads

$$S = S_B + \frac{1}{2} J \sum_{a \neq b} \int_0^\beta d\tau \; \tilde{S}_a \cdot \tilde{S}_b +$$

$$+ \frac{1}{2} \int_0^\beta d\tau d\tau' \sum_{ab} D_{ab}(\tau - \tau') \tilde{S}_a(\tau) \cdot \tilde{S}_b(\tau'),$$ \hfill (2)

in which $S_B$ denote Berry phase terms. $D_{ab}(\tau - \tau')$ is a retarded interaction, generated by integrating out all spins outside the plaquette. Higher order interactions are also induced in this process, which have been neglected in (2). Equivalently, one can view the rest of the lattice as an external bath which couples to the spins in the cluster through time-dependent external fields with a Gaussian correlator $D_{ab}$. The latter will be determined by a self-consistency requirement, which stipulates that the correlation functions on a plaquette calculated from the above action $(\chi_{ab}(\tau - \tau') = \frac{1}{2}(\tilde{S}_a(\tau) \cdot \tilde{S}_b(\tau))_S)$ involve the same self-energy as the entire lattice. This reads: $\chi_{ab}^T(i\nu_n) = J_{\Delta_{ab}} + D_{ab}(i\nu_n) + \mathcal{M}_{ab}(i\nu_n)$, where $J_{\Delta_{ab}} \equiv J(1 - \delta_{ab})$ is the matrix of nearest neighbor couplings on the cluster. Inserting this relation for the self-energy into (1) and imposing that $\sum_{\vec{q}} \chi_{ab}(\vec{q}) = \chi_{ab}$ leads to the final form of the self-consistency requirement

$$\chi_{ab}(i\nu_n) = \sum_{\vec{q}} \left[J(\vec{q}) - J\Delta + \chi^{-1}(i\nu_n) - D(i\nu_n)\right]^{-1}_{ab}.$$ \hfill (3)

We note that the matrix $J(\vec{q})_{ab} - J\Delta_{ab}$ involves only the exchange constants linking together different (uppointing) triangular plaquettes. In the limit where these inter-plaquette couplings vanish while the internal ones are kept fixed (decoupled triangles), our approach becomes exact since (1) implies $D_{ab} = 0$ and (2) reduces to the action associated with the Heisenberg model on a single triangle. We also note that the above DMFT equations can be derived from a Baym-Kadanoff formalism in which a functional of the correlation function and self-energy matrix is introduced in the form

$$\Omega[\chi, \mathcal{M}] = \frac{1}{2} \text{Tr} \ln[\mathcal{M} + J] - \frac{1}{2} \text{Tr}[\chi \mathcal{M}] + \Phi[\chi].$$ \hfill (4)

The stationarity conditions $\frac{\partial \Omega}{\partial \chi} = \frac{\partial \Omega}{\partial \mathcal{M}} = 0$ lead to the above equations when the exact functional $\Phi$ is approximated by its value for a single triangular plaquette. The free energy of the model can thus be calculated in the DMFT approach by inserting the self-consistent values of $\chi$ and $\mathcal{M}$ into (5). Alternatively, a functional of the correlation function only can be used, along the lines of Ref. [2].

In a phase with unbroken translational invariance, the matrix $\chi_{ab}(i\nu_n)$ in fact reduces to two elements: the local susceptibility $\chi_{aa} = \chi_{\text{loc}}$, and the nearest-neighbour one $\chi_{mn} = \chi_{ab}(a \neq b)$ (and similarly for $D_{ab}$). It is actually convenient to use the following linear combinations (proportional to the eigenvalues of the $\chi_{ab}$ and $D_{ab}$ matrices): $\chi_s = 3(\chi_{\text{loc}} + 2\chi_{mn})$, $\chi_m = 3(\chi_{\text{loc}} - \chi_{mn})$, $D_s = \frac{1}{2}(D_{\text{loc}} + 2D_{mn})$, $D_m = \frac{1}{2}(D_{\text{loc}} - D_{mn})$. Introducing the corresponding self-energies $M_s(i\nu_n) \equiv 3/\chi_s(i\nu_n) - 3D_s(i\nu_n)$, $M_m(i\nu_n) \equiv 3/\chi_m(i\nu_n) - 3D_m(i\nu_n)$, straightforward but tedious algebra allows us to recast (5) in the form of two scalar equations:
Here, $\epsilon$ stands for $\cos q_1 + \cos q_2 + \cos q_3$, where the $q_i$ are components of $\vec{q}$ along the three directions of the triangular lattice ($q_1 + q_2 + q_3 = 0$), and $\rho(\epsilon)$ is the corresponding density of states. The $\vec{q}$-summation in (4) simplifies into an $\epsilon$-integration in (6).

Equations (4,5,6) entirely define the plaquette-DMFT approach introduced here and one could embark at this stage into a numerical determination of the two key dynamical quantities $\chi_{\text{loc}}(\tau), \chi_{\text{m}}(\tau)$ using, e.g., a generalisation of the quantum Monte Carlo algorithm recently introduced in [13] in the context of quantum spin-glasses. Instead, we shall gain further insight into the problem by making use of a projection onto the low-energy sector of the Hilbert space [12].

The low-energy projection of the action (2) is then easily obtained as

$$S_{\text{low}} = \frac{1}{3} S \left[ 1 - 2 \omega^1 - a T^\gamma - 2 \omega^a - 1 T^+ \right]$$

(6)

The low-energy projection of the action (3) is then easily obtained as

$$S_{\text{low}} = S_B + \int_0^\beta d\tau d\tau' \left[ \frac{1}{2} D_s(\tau - \tau') + D_m(\tau - \tau') \vec{T}(\tau) \cdot \vec{T}(\tau') \right] \vec{S}(\tau) \cdot \vec{S}(\tau')$$

(7)

where $D_s$ and $D_m$ are as defined earlier. It turns out that $\chi_s$ and $\chi_m$ have simple interpretations as the “total spin” and “mixed” correlation functions:

$$\chi_s(\tau - \tau') = \frac{1}{3} \langle \vec{S}(\tau) \cdot \vec{S}(\tau') \rangle$$

$$\chi_m(\tau - \tau') = \frac{1}{3} \langle \vec{S}(\tau) \cdot \vec{S}(\tau') \vec{T}(\tau) \cdot \vec{T}(\tau') \rangle$$

(8)

To solve the local quantum problem defined by (6), we use an approximate technique that has proven successful in recent studies of quantum spin-glass models [10] and of impurities in quantum antiferromagnets [13].

We introduce two doublets of spin-1/2 fermions $(s_1, s_\downarrow)$ and $(t_1, t_\downarrow)$ subject to the constraints: $s_1^+ s_\downarrow = t_1^+ t_\downarrow = 1$ in order to represent the operators $\vec{S}$ and $\vec{T}$ as: $S^+ = s_1^+ s_\downarrow, S^- = s_\downarrow^+ s_\uparrow, S_z = (s_1^+ s_\downarrow - s_\downarrow^+ s_\uparrow)/2$; $T^+ = t_1^+ t_\downarrow, T^- = t_\downarrow^+ t_\uparrow$. The interacting fermion problem corresponding to (6) is then solved in the self-consistent Hartree approximation. Also, the local constraint is approximated by its average (the associated Lagrange multiplier is zero due to particle-hole symmetry). Introducing the Green’s functions $G_s$ and $G_t$ for the two fermion fields and defining self-energies by $G^{-1}_s(i\omega_n) = i\omega_n - \Sigma_s(i\omega_n)$, $G^{-1}_t(i\omega_n) = i\omega_n - \Sigma_t(i\omega_n)$ (with $\omega_n = (2n + 1)\pi/\beta$ a fermionic Matsubara frequency), this yields the imaginary-time equations:

$$\Sigma_s(\tau) = -\frac{3}{2} \left[ 2 \right] D_s(\tau) G_s(\tau) + 3 D_m(\tau) G_s(\tau) G_t(\tau) G_t(\tau), \Sigma_t(\tau) = 3 D_m(\tau) G_t(\tau) G_s(\tau) G_t(\tau).$$

(9)

Then the local problem (for a given bath $(D_s, D_m)$) reduces to two coupled non-linear integral equations. The correlation functions are given by

$$\chi_s(\tau) = -\frac{1}{2} \frac{D_s(\tau)}{G_s(\tau)} \frac{G_s(\tau)}{G_s(\tau)}, \chi_m(\tau) = 2 G_s(\tau) G_s(\tau) G_t(\tau) G_t(\tau).$$

(10)

In practice, we use the following algorithm: we start with an initial guess for the bath $(D_s, D_m)$ and obtain the Green’s functions by iteration of eq. (6). We can then calculate the susceptibilities from (10), as well as the self energies $\Sigma_{s,m}$. Inserting the latter into (6) yields new $\chi_{s,m}$ and, in turn, the new baths $D_{s,m}^{\text{new}}(i\nu_n) = -\Sigma_{s,m}(i\nu_n)/3 + 1/\chi_{s,m}(i\nu_n)$.

We now discuss our findings when solving these coupled equations numerically. The first key observation is that a paramagnetic solution can be stabilized down to the lowest temperature we could reach, with no sign of long-range ordering intervening. Long-range order is associated with a diverging eigenvalue of $\chi_{ab}(\vec{q})$ for some $\vec{q}$ and hence, within our scheme, to a vanishing denominator in (6), which we never observe. In Fig. 2, we display our results for the temperature dependence of the local (i.e on-site, or $\vec{q}$-integrated) susceptibility $\chi_{\text{loc}}(T)$, $\chi_{\text{loc}}(T) d\tau$. At high temperatures $\chi_{\text{loc}} \propto 1/T$ obeys Curie’s law. Below $T \approx 0.5 J$, the effective Curie constant decreases with decreasing $T$, indicating gradual quenching of the local moment. However, $\chi_{\text{loc}}$ itself continues to increase as the temperature is lowered. In the inset of Fig. 2, we display the effective exponent defined by $\alpha(T) = -\ln \chi_{\text{loc}}/\ln T$. A transient regime, corresponding to a slowly decreasing $\alpha$, is apparent and extends over...
more than two decades. At the lowest temperatures, $\alpha$ appears to saturate to a value $\alpha \simeq 0.5$.

![Graph](image)

**FIG. 2.** Main plot: $\chi_{loc}$ obeys Curie law ($\propto 1/T$) at high temperature, but diverges more slowly than $1/T$ at lower temperature. Inset: Evolution of the effective exponent $\alpha$ from high to low temperature when fitting $\chi_{loc}(T)$ to $1/T^\alpha$. ($T$ is in units of $J$ in all figures.)

For different (low) temperatures, $\chi_{loc}(\tau)/\chi_{loc}(\beta/2)$ plotted as a function of $\tau/\beta$ collapse on a single curve, well described by $[\sin(\pi \tau/\beta)]^{-0.5}$ (solid curve).

![Graph](image)

**FIG. 3.** Evolution of the effective exponent $\alpha$ at different (low) temperatures, with $1/\chi_{loc}(T)/\omega_{loc}$ increasing at low temperatures in our approach, down to an unphysically low energy scale (of order $J/100$). In fact the triplet gap for the $S = 1/2$ kagome QAF is known to be quite small (of order $J/20$), so our description of the spin dynamics should be valid down to a rather low temperature scale. The power-law behaviour of the spin correlations and the low-temperature increase of the susceptibility agree well with experimental findings on SCGO above its freezing temperature at $T = 4K$. Since this is a $S = 3/2$ system, it is conceivable that higher spin extends the range of applicability of our approach even further. A promising application of our approach is the pyrochlore lattice: the natural plaquette is a tetrahedron, with a twofold degenerate singlet ground state, so that the very low-energy singlet sector should be accessible within our approach. In future work we plan to address these issues, and also study the low-temperature thermodynamics by solving the self-consistent local problem using exact numerical techniques.

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