Dynamical Correlations of the Kagome $S = 1/2$ Heisenberg Quantum Antiferromagnet

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We determine dynamical response functions of the $S = 1/2$ Heisenberg quantum antiferromagnet on the kagome lattice based on large-scale exact diagonalizations combined with a continued fraction technique. The dynamical spin structure factor has important spectral weight predominantly along the boundary of the extended Brillouin zone and energy scans reveal broad response extending over a range of $2 \sim 3J$ concomitant with pronounced intensity at lowest available energies. Dispersive features are largely absent. Dynamical singlet correlations – which are relevant for inelastic light probes – reveal a similar broad response, with a high intensity at low frequencies $\omega/J \lesssim 0.2J$. These low energy singlet excitations do however not seem to favor a specific valence bond crystal, but instead spread over many symmetry allowed eigenstates.

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Introduction

The $S = 1/2$ Heisenberg quantum antiferromagnet (AFM) on the kagome lattice is a key model for our theoretical understanding of highly frustrated quantum magnets in two spatial dimensions. In contrast to other model systems, such as the checkerboard magnet, it continues to hide the true nature of its ground state despite a longstanding effort. On the experimental side many kagome like materials were discovered and characterized over the years, and some of them seem to get close to the goal of an experimental realization of a perfect Heisenberg $S = 1/2$ kagome system. Recently the synthesis of powder samples of the Herbertsmithite ZnCu$_3$(OH)$_6$Cl$_2$ [1] and the subsequent experimental investigations sparked a new wave of theoretical interest in this long standing problem [2].

So far most of the theoretical studies focused on ground state properties, discussing various possible phases, such as valence bond solids [3], gapped spin liquids of different kinds [4, 5, 6], and also stable critical phases [7, 8]. Much less attention however has been paid to the precite nature and form of the low-energy excitations visible in frequency resolved probes such as inelastic neutron scattering or light scattering techniques. In the present Letter we fill this void and present a detailed numerical study of the dynamical response of $S = 1/2$ kagome systems in the spin triplet and singlet channels and formulate predictions to be tested in inelastic scattering experiments.

We study the $S = 1/2$ Heisenberg quantum antiferromagnet on the kagome lattice, governed by the Hamiltonian:

$$H = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$$

where $J > 0$ is the antiferromagnetic nearest neighbor exchange coupling. Our results are based on large-scale exact diagonalizations of up to $N = 36$ spins, supplemented by the continued fraction method [9] for dynamical correlations functions. For the dynamical quantities we performed 500 (3000) iterations in Hilbert spaces of dimensions $3.8 \times 10^8$ ($4.5 \times 10^9$) for the spin and singlet dynamics respectively.

A lot of our present understanding of $S = 1/2$ systems is based on a series of exact diagonalization studies [10], which convincingly showed the absence of magnetic order, and revealed a puzzlingly high density of low-energy singlet and triplet excitations, a so far unique phenomenon. In the following we explore how the huge density of low-lying excitations affects experimentally relevant response functions.

Static spin response

The static spin structure factor is given by the following expression:

$$S^z(Q) = \frac{1}{\sqrt{N}} \sum_j e^{-iQr_j} S^z_j,$$

$$S(Q) = \langle S^z(-Q)S^z(Q) \rangle,$$  \hspace{1cm} (2)

where the wave vector $Q$ is not restricted to lie in the first Brillouin zone (BZ).

The numerically determined static spin structure factor for the 36 sites sample is shown in Fig. 1, as an intensity plot covering multiple BZs [right hand side, plot (1)], and along a path in the extended BZ [plot (2)]. The response is strong and broad along the zone boundary of the extended BZ, revealing the short ranged nature of the antiferromagnetic correlations [10]. In the $N = 36$ groundstate there are additional small peaks at the point ($g$), a feature which is also reported in a recent DMRG study [6]. Comparison with a $N = 24$ sample (not shown) confirms that the broad response along the boundary of the extended BZ is a generic, size-independent feature, characterizing a state with spin correlations which are decaying rapidly beyond the nearest neighbor sites. To contrast this result with a magnetically ordered state we show the static response in the $q = 0$ [Fig. 1(3)] and $\sqrt{3} \times \sqrt{3}$ [Fig. 1(4)] states, which have been obtained using a strong $J_2$ coupling with the appropriate sign.

Dynamical spin structure factor

The energy and momentum dependence of the dynamical structure factor:

$$S(Q, \omega) = -\frac{1}{\pi} \text{Im} \langle S^z(-Q) \frac{1}{\omega - (H - E_{GS}) + i\eta} S^z(Q) \rangle,$$ \hspace{1cm} (3)
FIG. 1: (Color) Dynamical spin structure factor of the $N=36$ sample. The eight panels display frequency scans $S(Q,\omega)$ ($\eta = 0.02J$) at labeled wavevectors $Q$ in the extended Brillouin zone shown in the lower right center. Note that the intensity scales differ among the different panels. The $\Gamma$ point has no weight and is not shown. The blue vertical lines show the pole location and intensity of the continued fraction. The vertical dotted magenta line denotes the finite size spin gap in the corresponding momentum sector. The dashed red line marks the position of the first frequency moment $\bar{\omega} = \int d\omega \omega S(Q,\omega)/S(Q)$. In the rightmost column the static spin structure factor of the pure Heisenberg model on the kagome lattice is shown, as an intensity plot (1) and along the path $\Gamma - (e) - (g) - \Gamma$ (2). The static structure factor for the $q = 0 (3)$ and $\sqrt{3} \times \sqrt{3}(4)$ Néel order states induced by appropriate second neighbor couplings are also displayed.

is directly relevant for inelastic neutron scattering (INS) experiments and therefore a quantity of central interest. In magnetically ordered systems we expect to see dispersive, long-lived spin waves [11], while one-dimensional systems in appropriate regimes can display spinon continua with a rich structure [12].

Our numerical results for the $N = 36$ kagome lattice are presented in the left part of Fig. 1. The shaded panels display an energy scan at the wave vector indicated by the panel position and its label, referring to specific points in the extended BZ. Each panel displays the broadened ($\eta = 0.02J$) spectral function (black line), the locations and weights of the poles of the continued fraction expansion (blue vertical lines), the finite size spin gap in the corresponding momentum sector (dotted vertical line), and the first frequency moment $\bar{\omega}(Q) = \int d\omega \omega S(Q,\omega)/S(Q)$ (dashed vertical line).

Consistent with the static structure factor [by virtue of the sum rule $S(Q) = \int d\omega S(Q,\omega)$], the dynamical spin response function concentrates essentially along the boundary of the extended BZ. The main feature of this system is the stretching of the magnetic response in each $Q$-sector on a very large number of excited states spanning a large bandwidth of $2 \sim 3J$, starting immediately above the (finite-size) gap. Furthermore there seems to be a pronounced enhancement of the intensity at small $\omega$. The different spectral functions look rather similar, suggesting an approximate factorization $S(Q,\omega) \sim S(Q) \times f(\omega)$, at least at intermediate and high $\omega$. The overall picture is definitely quite different from the spectrum of a Néel ordered system on the same system size, where an overwhelming part of the spectral weight is carried by very few poles in each $Q$-sector associated to the Bragg peak and the one-magnon modes respectively [13]. Still at some wave vectors the lowest pole carries significant weight especially at $(g)$. The origin of this feature remains to be elucidated but could potentially come from an algebraic divergence in one scenario [8] or from triplon excitations on top of (remnants of) a valence bond crystal in a different scenario [14].

In order to address finite-size effects we present two spectral functions at the wave vectors $(g)$ and $(i)$ for $N = 24$ and 36 spins in Fig. 2(a) and (b). The characteristic width in energy as well as the prominent response at low $\omega$ for wave vector $(g)$ are clearly stable with respect to finite size effects. In panels (c) the local dynamical spin correlation function $S_{\text{loc}}(\omega) \propto \int dQ S(Q,\omega)$ is shown for different system
Local Dimer Dynamics(ω)

Accumulated S(ω)

S(ω)0.2

0.4

0.6

0.8

ω/J

0

0.05

0.1

0.15

ω/J

0

0.2

0.4

0.6

0.8

N=36, Kagome

N=32, Checkerboard

N=26, Square

Domain Walls Multi-Triplon Continuum

FIG. 3: (Color) Dynamical singlet fluctuations [Eq. (4)] for three different systems. Main plot: kagome lattice. Upper inset: Checkerboard lattice with a plaquette-like valence bond crystal ground state. Lower inset: Unfrustrated square lattice exhibiting Néel order. The plotted quantity represents well the qualitative features of the Raman response of the three systems.

sizes, again highlighting the stability of the overall shape of the spectral function. Finally panel (d) presents the cumulative spectral weight as a function of ω/J. All spectral functions have been broadened using η = 0.05J.

Effect of impurities We have studied the influence of a low concentration of nonmagnetic impurities on the spin dynamics by depleting a N = 27 sample by one site. The averaged dynamical spin response closely resembles Fig 2(c) with an additional resonance-like feature at ω ∼ J due to the strong singlet forming on the two bonds next to the vacant site [15]. Due to its local nature, this feature is expected to be generically present in S(Q,ω) as well.

Singlet fluctuations In order to assess the importance of the abundant number of low energy singlet excitations for optical probes and to investigate the tendency towards valence bond crystal ordering, we study the local dynamical fluctuations of a nearest neighbor dimer operator:

\[ D_{i,j} = S_i \cdot S_j - \langle S_i \cdot S_j \rangle \]
\[ D_{i,j}(\omega) = -\frac{1}{\pi} \text{Im} \langle D_{i,j} \rangle \frac{1}{\omega - (H - E_{GS}) + i\eta} D_{i,j} \] (4)

The interest in this quantity is twofold. First Eq. (4) is closely related to the Raman or RIXS response of a spin system and thus reveals the qualitative features of the inelastic light scattering response. And second, we expect a spontaneous translational symmetry breaking due to dimerization to show up as an important ω → 0 contribution.

The fluctuation spectrum for the N = 36 kagome system is shown in Fig. 3, where a broad response from the lowest singlet up to energies ∼ 4J is seen together with a strong increase of the response towards the lowest energies. The kagome result can be compared to the response of the Heisenberg model on a checkerboard lattice (upper inset in Fig. 3) and the square lattice (lower inset in Fig. 3), where in both cases the physical origin of the response is essentially understood. On the Néel ordered square lattice the dimer-dimer response is assigned to the two-magnon continuum with a maximum strength around 3J. This is obviously very similar to the Raman response of the square lattice Heisenberg AFM [16]. On the checkerboard lattice with its plaquette valence bond crystal ground state [17] the dimer-dimer response function shows different frequency domains: a single low-lying peak (shaded in red) originating from the valence-bond symmetry breaking partner of the ground state, followed by two domains at non zero-frequency, first a range of singlet excited levels (shaded orange) which have been convincingly explained as valence bond crystal domain-wall excitations [17, 18] and a second range ω ≳ J corresponding to a multi-triplon continuum [17] (shaded in brown). It is clear from this comparison that the kagome lattice does not show typical valence bond crystal characteristics exemplified by the checkerboard magnet. Still the response on several levels up to ω ≲ 0.2J seems to be particular strong. However this response spreads on many low lying levels in any symmetry sector which can be excited by the dimer-dimer operator. We do not see any
clear precursor of a specific spatial symmetry breaking pattern when comparing the excited levels to different valence bond crystal symmetry predictions summarized in Ref. [19]. Possible reasons are: i) the absence of VBC order (see Ref. [6] for a similar conclusion) or ii) a very weak ordering with a very large unit cell which has not still emerged from competing orders.

**Comparison to theoretical proposals** Both the spin and the dimer dynamical fluctuations of a $S = 1/2$ kagome system are intrinsically broad, and are not easy to reconcile with the excitation spectrum scenarios for the various proposed ground states. For example the dynamical spin correlation functions lack the characteristic coherent triplon excitations, which are expected on top of a valence bond crystal [4]. A critical spin liquid would possibly have similar low-energy response at low energies. This fluctuating background provides also a natural explanation for the reported absence of quasiparticles in a doped kagome system [25]. Building on these results it will be interesting to understand the evolution of the dynamical response at finite temperature as well as the effect of a magnetic field. Given that many experimental kagome systems have some (small) Dzyaloshinsky-Moriya interactions their influence on the dynamical response is also worth studying.

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