Spin excitation spectrum of high-temperature cuprate superconductors from finite cluster simulations

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Abstract
A cluster of spins 1/2 of a finite size can be regarded as a basic building block of a spin texture in high-temperature cuprate superconductors. If this texture has the character of a network of weakly coupled spin clusters, then spin excitation spectra of finite clusters are expected to capture the principal features of the experimental spin response. We calculate spin excitation spectra of several clusters of spins 1/2 coupled by Heisenberg interaction. We find that the calculated spectra exhibit a high degree of variability representative of the actual phenomenology of cuprates, while, at the same time, reproducing a number of important features of the experimentally measured spin response. Among such features are the spin gap, the broad peak around \( \hbar \omega \approx (40–70) \text{ meV} \) and the sharp peak at zero frequency. The latter feature emerges due to transitions inside the ground-state multiplet of the so-called ‘uncompensated’ clusters with an odd number of spins.

Keywords: magnetic susceptibility, high-temperature superconductivity, neutron scattering experiments, cuprates

1. Introduction

Neutron scattering experiments in high-temperature cuprate superconductors reveal intricate patterns of magnetic spin response [1, 2]. Recently some of these findings were corroborated by the resonant inelastic x-ray scattering (RIXS) experiments [3]. A broad range of theoretical approaches of varying degree of sophistication, including those based on Hubbard [4–6], \( t - J \) [7–10], Heisenberg [11–15], spin-fermion [16] models and their extensions have been employed in order to reproduce this spin response. At present, however, there is no consensus about the starting set of assumptions to describe the available phenomenology in cuprates. Here one faces dilemmas between the pictures of itinerant and localized spins, between including inhomogeneous spin textures at the level of model assumptions or obtaining these textures dynamically from the spin susceptibility of the homogeneous parent state. When inhomogeneous textures are assumed, one has a choice of either stripe or checkerboard patterns, or more disordered ‘Swiss-cheese’-type of textures [17, 18]. In the latter case, one expects that the antiferromagnetic order is retained locally within finite spin clusters (domains, puddles etc), at least approximately, while inter-cluster correlations fade away with doping. In general, irrespective of the initial set of assumptions, one can reasonably expect that the spin response at sufficiently high frequencies for infinite systems and for finite parts of these systems would be approximately the same. If, however, the spin texture has the character of a network of weakly coupled spin clusters, the cluster calculations can also capture important

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\(^{3}\) See the supplemental material in arXiv:1712.09979
experimentally observed features at low frequencies such as the onset of a spin gap. Such a texture can, indeed, emerge as a possible realization of Coulomb-frustrated phase separation [19, 20]. In this work, we investigate spin responses of finite clusters of spins 1/2 described by the Heisenberg model, in an attempt to test whether the experimental phenomenology can be understood from a unified perspective of a finite-cluster simulations. We find that such an approach is indeed quite promising.

We start in the next section with a brief overview of the main features of spin response observed in cuprates along with related theoretical concepts and considerations. In section 3, we formulate our cluster model and discuss its general properties. Several clusters are considered, with sizes and shapes motivated by theoretical considerations and experimental data. In section 4, we present the results of numerical simulations and discuss their relation to the experimental data. The simulated magnetic response from these clusters exhibits several features observed experimentally across a wide range of cuprate compounds. Discussion and conclusions are finally presented in section 5.

2. Main features of spin response in cuprates

We consider the spin excitation spectrum probed by neutron scattering experiments. This technique allows one to measure the imaginary part of the dynamical susceptibility $\chi''(Q, \omega)$. In the low-temperature limit, it is defined as [2]

$$\chi''(Q, \omega) = \sum_{\alpha, \beta} \left( \delta_{\alpha\beta} - Q_\alpha Q_\beta / Q^2 \right) \times \frac{1}{2\pi} \int \frac{d\varepsilon}{\varepsilon} \sum_{r' r} e^{iQ(r - r')} \langle S_r^\alpha(0) S_{r'}^\beta(t) \rangle, \quad (1)$$

where $S_r$ is the operator of the spin 1/2 in the lattice site with the coordinate $r$ and the quantum mechanical average, $\langle S_r^\alpha S_{r'}^\beta \rangle$, is taken over the subspace of ground states. We put $\hbar = 1$. One can also define the $Q$-integrated local susceptibility $\chi''(\omega)$ as

$$\chi''(\omega) \equiv \int dQ \chi''(Q, \omega). \quad (2)$$

A large body of experimental work has been done on the neutron scattering in high-temperature cuprate superconductors (see e.g. [1, 2] for reviews). Detailed experimental data are available for a number of compounds, for which sufficiently large single-crystal samples can be produced. Below we list the principal features of these data. For convenience of the reader, we reproduced several relevant experimental plots in the reference provided in footnote 3. We refer to these plots, figures S1 and S2, on several occasions below.

2.1. Spin gap versus a peak at $\omega \approx 0$

The spin response around zero frequency comes in two variations. On the hole-doped side, underdoped lanthanum cuprates exhibit a narrow peak at zero frequency followed by a frequency range with suppressed response and then one or more somewhat broader peaks [24, 25] (see figures S1(a) and (e) in footnote 3), while optimally doped and overdoped lanthanum cuprates [26, 27] as well as other not too underdoped cuprates [28–32] generally exhibit a spin gap, defined as a frequency threshold below which $\chi''(\omega)$ vanishes (see footnote 3, figures S1(b)–(d), (f) and (g)). This spin gap varies from a few meV to tens of meV. In the case of optimal doping, it appears to correlate with the critical temperature [2]. In electron-doped cuprates, a peak at $\omega \approx 0$ was also reported in [33]. However this peak is possibly separated from $\omega = 0$ by a small gap, see figure S1(h) in footnote 3. It should be noted that, even in the compounds where spin gap is apparently absent, the magnetic response at low frequencies is very different from that of parent antiferromagnets, where it is dominated by spin waves [13, 34].

From the theoretical standpoint, a spin gap or its absence appears to be a rather delicate issue. While the gap is absent in a Heisenberg model for spins 1/2 both on a square lattice and in a one-dimensional chain, it can appear in spin ladders with even number of legs [35]. This supports the idea that spatial inhomogeneities can be responsible for the gap. On the other hand, the gap in spin ladders is not particularly robust and can vanish for ladders with next-nearest and four-spin interactions [36].

2.2. Hourglass and wine glass structure of $\chi''(Q, \omega)$

In doped cuprate superconductors the antiferromagnetic peak splits in a way which often, but not always, gives rise to the celebrated hourglass structure—a feature common for a variety of compounds [1, 2], see figure S2(a) in footnote 3. An alternative to the hourglass is a Y-shaped structure also known as the ‘wine glass’ which has been observed in a slightly underdoped $\text{HgBa}_2\text{CuO}_4$, see figure S2(b) in footnote 3. We note, however, that the upper branch of the hourglass is similar to the upper branch of the wine glass and therefore constitutes a more universal feature. This upper branch also appears in spin responses of various theoretical models, such as spin ladder and Hubbard models [4, 5, 24, 37]. As for the lower branch, its theoretical description requires an accurate treatment of the lower-energy physics (for example, interstripe interactions in the stripe paradigm) and thus is more difficult to justify [8, 9, 11, 13, 38].

2.3. Broad peak in $\chi''(\omega)$ at $\omega_0 = (40–70) \text{ meV}$

This feature is rather universal for hole-doped cuprates. It is observed in all cases shown in figure S1 footnote 3, except for the one shown in figure S1(c) footnote 3 (the overdoped $\text{La}_2\text{−}_x\text{Sr}_x\text{CuO}_4$ [27]). When this peak exists, its frequency $\omega_0$ corresponds to the waist of the hourglass or the bottom of the wine glass. Both the origins of this peak and its implications for the high-temperature superconductivity attracted a lot of theoretical and experimental attention [38]. From the theoretical point of view this peak often emerges as another facet of the spin gap [4, 5, 9, 11, 28, 38].
2.4. Sharp low-frequency peak in $\chi''(\omega)$

This peak has been observed in underdoped, optimally doped and overdoped La$_{2-x}$Sr$_x$CuO$_4$ [25–27, 39] at $\omega = (7–18)$ meV, see figures S1(a)–(c) footnotes 3. Its apparent counterpart appears in La$_{1.825}$Ba$_{0.125}$CuO$_4$ [24] at $\omega = 41$ meV and, possibly, in YBa$_2$Cu$_3$O$_{6+\delta}$ [31] at $\omega = 33$ meV, see figures S1(e) and (d) footnote 3, respectively. It is an open question whether all these peaks share the same origin.

Concluding this review, we would like to mention that one notable dilemma unresolved so far is the dimensionality of spin modulations in cuprates, static or dynamic, and if and when they exist. Two options are commonly discussed: a one-dimensional striped spin texture (see e.g. [40] and references therein) or a two-dimensional checkerboard texture (see e.g. [41, 42] and references therein). Attempts have been made to discriminate between these options, in particular, by analyzing experimentally measured splitting of the antiferromagnetic peak in $\chi''(Q, \omega)$. However, no consensus on this matter has emerged up to date [21, 43–46].

3. Model and preliminary considerations

3.1. Theoretical model

We consider five clusters of spins 1/2 shown in figure 1 with the total number of spins, $N$, equal to 5, 9, 12, 13 and 16. Each cluster is a piece of a square lattice where spins are coupled by the nearest-neighbour Heisenberg interaction described by the Hamiltonian

$$H = J \sum_{\langle \mathbf{r}, \mathbf{r}' \rangle} \mathbf{S}_\mathbf{r} \cdot \mathbf{S}_{\mathbf{r}'} ,$$

where $\langle \mathbf{r}, \mathbf{r}' \rangle$ is a pair of nearest neighbours, $J$ is the coupling constant. The value of $J$ in parent cuprates lies in the range (100–150) meV [2]. For numerical estimates we take the value $J = 120$ meV.

It should be noted that, since we study isolated spin clusters, no periodic boundary conditions are imposed. The Fourier transform of a non-periodic function is continuous rather than discrete. We calculate this Fourier transform (as defined by equation (1)) numerically with a sufficiently small momentum resolution appropriate for the resulting continuous spectrum.

The linear sizes of the clusters considered are (2–4) lattice constants. These sizes roughly correspond to typical $Q$-scale of features of the experimentally observed $\chi''(Q, \omega)$. Similar characteristic sizes of modulated spin superstructures appear in a number of theoretical proposals involving, in particular, stripes [40] and checkerboards [21, 41].

The excitation spectrum of each cluster, if described only by the Hamiltonian (3), consists of a finite number of discrete frequencies. This means that the magnetic susceptibilities (1) and (2) are reduced to sums of $\delta$-functions, $\delta(\omega - \Omega_i)$, where $\Omega_i$ is the the frequency of the transition between two discrete energy levels. In reality, various effects not included in the above simple model would broaden these sharp spectral lines. These effects include fluctuations of the shape and the size of the clusters, charge carriers hopping on and off the clusters and intercluster interactions. Therefore, we introduce a phenomenological Lorentzian broadening with half-width $\Gamma$ by substituting

$$\delta(\omega - \Omega_i) \rightarrow 1 \quad \frac{1}{\pi \Gamma} \quad \frac{1}{(\omega - \Omega_i)^2 + \Gamma^2} \quad .$$

We estimate the value of $\Gamma$ as the half-width of most fine details in the experimentally measured magnetic response. Unlike explicitly stated otherwise, we choose $\Gamma = 0.1J = 12$ meV, in line with experimental data obtained for La$_{2-x}$Sr$_x$CuO$_4$ [26] and YBa$_2$Cu$_3$O$_{6+\delta}$ [31]. It should be kept in mind, however, that $\Gamma$ can substantially vary from compound to compound.

Few remarks are now in order. (i) The numerical analysis of finite cluster was employed previously as an approximate method for accessing the spin response of infinite two-dimensional spin systems (see e.g. [47, 48]). With such a goal in mind it was natural to choose periodic boundary conditions. The agenda of the present work is different in the sense that we assume that real spin clusters possibly exist in a sea of itinerant electrons. Therefore we focus on the implications of the finiteness of these clusters. In particular, we use open boundary conditions. The difference between periodic and open boundary conditions can be dramatic for small clusters, especially for uncompensated ones (defined and discussed in the next subsection).

(ii) It is known [34] that, in order to accurately describe spin excitations in undoped parent cuprate compounds, one needs to supplement the nearest-neighbour Heisenberg coupling by next-nearest-neighbour and four-spin ring exchange couplings. While accounting for these couplings is important for quantitative description of spin excitations, we believe that including them in our analysis would exceed the accuracy of our basic assumption that the spin cluster consists of localized spins. Therefore keeping only the nearest-neighbour coupling in the Hamiltonian (3) should be sufficient for the qualitative and even semi-quantitative analysis of the spin-cluster scenario.

(iii) Finally, we comment on the absolute values of the intensities of the spin response. The experimentally measured absolute intensities are known to be significantly below the theoretical estimates based on sum rules in the Heisenberg or Hubbard models [1, 2, 49]. A careful analysis reduces but does not completely eliminate the tension between the theory and the experiment [49]. This problem can be straightforwardly resolved in the spin-cluster scenario by choosing the concentration of clusters in the sea of itinerant electrons sufficiently small to satisfy the experimentally measured absolute intensities. For this reason we present the spin susceptibilities of clusters up to an arbitrary normalization factor.

3.2. Lieb–Mattis theorem and the spin of the ground state

The Lieb–Mattis theorem [50] constrains the total spin $S_{\text{gs}}$ of the ground state of a spin cluster. It applies to bipartite spin lattices with the nearest-neighbour Heisenberg interaction (3), i.e. lattices which can be divided into two sublattices $A$ and $B$,
such that spins in each pair of nearest neighbours belong to different sublattices. The theorem states that

\[ S_{\text{gs}} \leq |S_A - S_B|, \]

where \( S_A \) and \( S_B \) are the maximal total spins of sublattices \( A \) and \( B \), respectively. These maximal spins can be expressed as \( S_A = \frac{1}{2}N_A \) and \( S_B = \frac{1}{2}N_B \), where \( N_A \) and \( N_B \) are the numbers of lattice sites in the respective sublattices.

We divide all clusters considered into two categories: compensated clusters with \( N_A = N_B \), and uncompensated ones, with \( N_A \neq N_B \), see Figure 1. The total spin of the ground state of a compensated cluster is always zero due to the Lieb–Mattis theorem. In contrast, the total spin of the ground state of an uncompensated cluster can be nonzero.

It should be noted that, for finite clusters, the Lieb–Mattis theorem is also applicable in the presence of not too large next-nearest-neighbour and four-spin ring exchange couplings (see [50, 51]). For this reason, the hierarchy of energy levels with different total spin and the structure of transitions (see below) dictated by the theorem is robust.

Another remark concerns boundary conditions. If we were to use the periodic boundary conditions, the compactification of the cluster with an odd number of spins would destroy the division of a lattice into sublattices and thus makes the Lieb–Mattis theorem inapplicable.

### 3.3. Selection rules

The spin susceptibility measured in neutron scattering experiments obeys the two selection rules: (i) the allowed transitions must satisfy the condition \( |\Delta S| \leq 1 \), where \( \Delta S \) is the difference between the total spins of the final and initial states; (ii) transitions between states both having zero total spin are forbidden.

### 4. Magnetic response of compensated and uncompensated clusters

#### 4.1. Compensated clusters

We consider two compensated clusters with \( N = 12 \) and \( N = 16 \), see Figure 1. The numerically calculated magnetic responses of these two clusters are presented in Figure 2. Below we discuss their main features.

#### 4.1.1. Spin gap

The ground state is a singlet separated from the first excited state by a gap. According to the selection rule (ii), there are no transitions at zero frequency. As a consequence, spin response at low frequencies vanishes.

#### 4.1.2. Singlet-to-triplet excitations at \( \omega = J/2 \approx 60 \text{ meV} \)

This is the most prominent feature seen in the plot of the integrated susceptibility \( \chi''(\omega) \). It originates from the transition between the spin-singlet ground state and the lowest spin-1 excited state. The value of this frequency fits well the typical frequency (40–70) meV of the broad peak discussed in Section 2. In the momentum plane, the peak is localized around \( Q = (\pi, \pi) \) without any sign of splitting.

#### 4.1.3. Features of \( \chi''(Q, \omega) \)

The upward dispersion of \( \chi''(Q, \omega) \) at frequencies \( \omega > \omega_0 \) is present for both clusters. The shapes of sections of \( \chi''(Q, \omega) \) at a given \( \omega \) are non-universal. The ring-shape pattern of \( \chi''(Q, \omega) \) seen in Figure 2 for some of constant-\( \omega \) sections is often observed in the experiments [1, 2]. When constant-\( \omega \) sections are not ring-shaped, their orientations depend on the orientations of the cluster’s boundaries and often exhibit \( \pi/4 \) rotations with changing \( \omega \).

We note that, as illustrated in Figure 1, the cluster with \( N = 12 \) can approximate the basic building block of the spin-vortex checkerboard pattern proposed in [21–23].
coupling between these blocks is expected to be relatively small because of the smaller size of staggered spin polarizations at block’s boundaries. The corners of these blocks are also separated from each other by spin vortex cores where the staggered spin polarizations almost vanish.

4.2. Uncompensated clusters

Three uncompensated clusters were considered, with \( N = 5 \), \( N = 9 \) and \( N = 13 \), see figure 1. According to the inequality (5) imposed by the Lieb–Mattis theorem the total spin of the ground states of these clusters, \( S_{gs} \), does not exceed 3/2, 1/2 and 5/2, respectively. We find that, in fact, \( S_{gs} \) assumes the above maximal values, in line with [52].

We note here that the degeneracy of the ground state is taken into account in equations (1) and (2) by averaging over the ground state subspace. This procedure is equivalent to performing calculations at finite temperature \( T \) and then taking the limit \( T \to 0 \).

The numerically obtained magnetic responses for uncompensated clusters are presented in figure 3. Their main features are the following:

4.2.1. Response at \( \omega \simeq 0 \). Since the transitions between the components of the ground state multiplet are not forbidden by the selection rules, the magnetic response has a peak at \( \omega = 0 \). The higher the multiplet degeneracy, the stronger the peak intensity. In the momentum plane, the intensity
associated with this peak is localized around \( Q = (\pi, \pi) \) without splitting.

4.2.2. Finite-frequency peaks. The lowest finite-frequency peaks for clusters with \( N = 5 \) and \( N = 13 \) are located in the frequency range \((40–70) \text{ meV}\) which is consistent with the experimentally observed broad peak discussed in section 2. At the same time, the lowest finite-frequency peak of the cluster with \( N = 9 \) is located at a significantly higher frequency of about 100 meV.

4.2.3. Dispersion of \( \chi''(Q, \omega) \). The dispersion of varying shapes around \( Q = (\pi, \pi) \) is generally present away from the zero frequency. As in the case of compensated clusters, the shape of the sections of \( \chi''(Q, \omega) \) at a given \( \omega \) correlates with the orientation of the clusters.

4.3. Spin response from the mixture of different clusters

It is possible that in a real system more than one type of clusters contribute to the magnetic response. Magnetic response from a population of different clusters can be obtained by combining responses from individual clusters with corresponding weights. As an illustration, we combine in figure 4 the responses from two clusters—a compensated cluster with \( N = 12 \) and uncompensated cluster with \( N = 13 \).

5. Summary and discussion

Our findings exhibited in figures 2–4 show that the magnetic responses from finite clusters of spin 1/2, despite high variability, exhibit features strongly reminiscent of the response measured in cuprates. These features are:

(i) spin gap in compensated clusters,
(ii) zero frequency peak in uncompensated clusters,
(iii) pronounced broad peak at \( \omega = (40–70) \text{ meV} \) present in both compensated and uncompensated clusters.

The spin gap in compensated clusters has a rather robust origin, namely, the transition from the spin-singlet ground state to the lowest spin-triplet excited state. Therefore, the interpretation of this gap in terms of compensated spin clusters is quite realistic and competitive with other proposed interpretations, e.g. based on spin ladders [35]. The zero frequency peak in uncompensated clusters emerging due to the transitions within the ground-state multiplet is even more remarkable, given that such a feature in underdoped lanthanum cuprates is particularly difficult to explain within all kinds of popular infinite-lattice models. Finally, a pronounced broad peak at \( \omega = (40–70) \text{ meV} \) emerges naturally as a finite size effect if one assumes that the linear size of clusters is about four lattice constants—an assumption supported by experimental data as well as theoretical considerations. This further strengthens the merits of the cluster paradigm. We also note that all clusters exhibit upward dispersion of \( \chi''(Q, \omega) \) which is, however, typical also for a broad class of infinite lattice models [4, 5, 9, 11–13, 15, 35, 53, 54].

At the same time, there are several features of spin response in cuprates which are not seen in clusters. These are: (i) the downward dispersion below the broad peak at \( \omega_0 = (40–70) \text{ meV} \) (lower part of the 'hourglass'), and (ii) the sharp peak at low frequencies of about \((7–18) \text{ meV}\) seen in lanthanum cuprates. The above discrepancies are not surprising—a model of isolated clusters should not be expected to reproduce well all low-frequency features, because they can depend on intercluster interactions and on the interactions with itinerant charge carriers and phonons [55, 56]. We note, however, that the mercury family of hole-doped cuprates and the praseodymium family of electron-doped cuprates exhibit the ‘wine glass’ response [30] more consistent with our cluster calculations. Whether this is an indication of non-interacting clusters or a mere coincidence remains to be clarified.

To summarize, we analysed the magnetic spin response in cuprates on the basis of the assumption that it may be coming from a collection of spin-1/2 clusters. We demonstrated that this approach is quite promising—it provides simple physical interpretations for a number of common features of the cuprate magnetic response, including the spin gap and the zero frequency peak.

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