Does the Heisenberg Model Describe the Multimagnon Spin Dynamics in Antiferromagnetic CuO Layers?

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We compute the absorption spectrum for multimagnon excitations assisted by phonons in insulating layered cuprates using exact diagonalization in clusters of up to 32 sites. The resulting line shape is very sensitive to the underlying magnetic Hamiltonian describing the spin dynamics. For the usual Heisenberg description of undoped Cu-O planes we find, in accordance with experiment, a two-magnon peak followed by high energy sidebands. However the relative weight of the sidebands is too small to reproduce the experiment. An extended Heisenberg model including a sizable four-site cyclic exchange term is shown to be consistent with the experimental data.

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The starting point of many theories describing high-temperature superconducting cuprates is the undoped parent compound. A consistent description of this phase is of great importance since the usual approach is to “extend” this model to describe the doped phase like the $t$-$J$ model [1] or the spin fermion model [2]. Also the understanding of this phase, namely, the physical realization of a two-dimensional (2D) spin-1/2 quantum antiferromagnet, is a fundamental problem in itself.

It is usually assumed that the 2D Heisenberg model (HM),

$$H_{Hei} = \sum_{i,j} J_{ij} S_i \cdot S_j,$$

(1)

describes the physics of the stoichiometric materials [3] with $J_{ij} = J$ for the first nearest neighbors and zero otherwise.

In this work we show that infrared (IR) optical absorption spectra due to phonon assisted multimagnon excitations is very sensitive to the magnetic Hamiltonian. We find that the usual Heisenberg description is incompatible with IR experimental data when more than two magnons are involved. An extended Heisenberg model including further neighbor interactions in $J_{ij}$ and a four-spin cyclic-exchange (4SCE) term [4–10] is shown to explain the IR experiments. The same model has been shown to be consistent with other spectroscopic data [10].

The 4SCE was introduced by Takahashi [5] and by Roger and Delrieu on the present context, using a 4th order perturbative analysis [6]. It was also supported by exact diagonalization (ED) studies of a multiband-Hubbard model [7] describing the Cu-O planes. The main effect of this term in the Hamiltonian is to permute cyclically four spins on a plaquette.

Though small exchange interactions, going beyond the first neighbors are expected [11], the 4SCE term is not generally accepted in the literature [3]. Up to now there has not been any clear experimental way to rule out or confirm the presence of this term. In fact, the experimentally established [3] single-magnon spin-wave branch is rather insensitive, at low energies, to the presence of the 4SCE term [7,12].

Any noticeable effect of the 4SCE term is expected to occur at high energies which are accessible in optical experiments. One example is magnetic Raman (MR) light scattering [13]. The line shape has an asymmetric peak close to $3J$ due to two-magnon excitations and a shoulder at higher energy which is believed to arise from a four-magnon process [14]. The width of the two-magnon peak and the four-magnon shoulder are anomalous in the sense that they do not agree with a conventional interacting spin-wave theory description of the line shape [6–11,15,16]. Theoretical studies have attributed both anomalies to the presence of other terms in the Hamiltonian [6–10,15,16] including the 4SCE term [6–10]. Although the latter assignment is encouraging we argue that the analysis of the MR line shape is not conclusive (see Ref. [17]).

Another experiment, which probes the multimagnon response, is phonon assisted multimagnon light absorption (PAMLA) [18–23]. In this experiment an absorbed photon simultaneously creates a phonon and a multimagnon excitation. The absorption mechanism is well understood allowing one to make theoretical predictions on the nickelates [22(b)] (2D, spin 1) which were successfully corroborated [19,20]. Besides, the line shape was computed for spin-1/2 1D Cu-O systems [23] with great accuracy [22(d)]. The experimental line shape is reproducible even among different materials (see Fig. 1). All this puts the interpretation of the IR line shape on a firmer basis than the MR line shape [17] making it an ideal candidate to test models of the spin dynamics.

For the 2D cuprates the line shape has been measured in several materials [18–21]. In Fig. 1 we show the
where $a$ has been subtracted from the raw absorption data. We show the function using Eq. (2). Following Refs. [18,20] a linear background has been subtracted from the raw absorption data. We show the curve, implying that this experiment depends only on the dimensionless scale. All the data collapse to a unique line shape for three different cuprate compounds in a close to 3D case, since a to the charge transfer band and the low energy upturn is common CuO 2D layers. The high energy upturn is due explained by the presence of a two-magnon process. In addition strong sidebands appear -exciton will depend on details outside the Hamiltonian is used. within the PAMLA mechanism if the appropriate magnetic average energy $r$ to the same quantity in MR ($r_{\text{Raman}}$) for different system size and for the experiment.

The physics of the two-magnon peak and the sidebands can be understood with a simple argument [9,22]. If one approximates the ground state by the classical Néel state the effect of the $B^+$ operator is to flip two spins in nearest neighbor sites. The energy of this excitation is $3J$ which is close to the energy of the peak observed. In the HM this state is not an eigenstate and will mix with states with four, six, etc., spin flips. Consequently the spectral function will show sidebands at the energy of these excitations which in the Ising limit are $4J$, $5J$, etc. Since the sidebands involve more than two magnons they cannot be described in the two-magnon-interacting spin-wave theory computation of Ref. [22(a),(b)]. It is therefore important to avoid the spin-wave approximation and establish with an unbiased technique whether or not the sidebands can be described by the HM. To this purpose we have computed the spectrum using ED on finite clusters.

Figures 1 and 2 show the exact spectrum in different size clusters. Although the Heisenberg line shape has some structure at the energy of the sidebands the relative intensity is much smaller than the experimental one. Notice the similarity with the two-magnon spin-wave theory line shape (dashed line in Fig. 1).

To make a more quantitative comparison we have computed the cumulants [11],

$$\langle M_n \rangle = \int (\omega - \rho)^n I(\omega) d\omega / I_T$$

with $I_T = \int I(\omega) d\omega$ and $\rho = \int \omega I(\omega) d\omega / I_T$.

$M_2$ and $M_3$ measure the width and asymmetry of $I(\omega)$, respectively, ($M_1 = 0$). They are obviously very sensitive to the presence of the sidebands so it is natural to use them to characterize the line shape.

In Fig. 3 we show $M_2$ and $M_3$ and the ratio of the average energy $\rho$ to the same quantity in MR ($\rho_{\text{Raman}}$) for different system size and for the experiment.

An extrapolation to an infinite system confirms what Figs. 1 and 2 suggest: the HM alone cannot correctly describe this experiment. It is difficult to ascribe this to a failure of the PAMLA mechanism itself since, as we mention above, the mechanism has been successfully
tested in an isostructural system with spin one, namely, La$_2$NiO$_4$ [19,22(b)] and a system with the same spin (1/2) but lower dimensionality [22(d),23]. We therefore analyze a more realistic Hamiltonian to describe the spin dynamics.

The Hamiltonian with the 4SCE term reads $H = H_{\text{Hei}} + H_{\text{4S}}$ with

$$H_{\text{4S}} = K \sum_{<i, j, k, l>} (S_i \cdot S_j)(S_k \cdot S_l) + (S_i \cdot S_j)(S_k \cdot S_l) - (S_i \cdot S_j)(S_k \cdot S_l),$$

where $<i, j, k, l>$ stands for the sum over groups of four spins on a plaquette. This term can be shown to produce the cyclic permutation of the four spins on the plaquette plus ordinary two-spins exchanges of all the pairs of spins of the plaquette including the ones on the diagonals (see Refs. [9,25]). The parameter $K/J$ has been estimated using an ED mapping from a multiband Hubbard model [7] to be around 0.3.

In Fig. 2 we show the exact line shape in the HM and in the model with the 4SCE term in a 26 site cluster.

We notice a strong sensitivity of the spectra to the 4SCE term. The main effect is to transfer weight from the first peak to the sidebands. We already see in this limited size cluster that the agreement with the experimental data is improved. As a consequence of this transfer of spectral weight one sees an increase of moments (Fig. 3).

A fourth order perturbative analysis of the Hubbard model [10] generates further nearest neighbor exchange interactions, beyond the 4SCE term already considered.

FIG. 2. $I(\omega)$ in ED for a 26 site cluster and with the same parameter sets as in Fig. 3. We also show the experimental line shape in Sr$_2$CuO$_2$Cl$_2$ [18 (thick line)]. The intensities were scaled to coincide at the maximum of the HM spectrum. In order to have the maximum of all the spectra at the same $\omega$ we have used different values of $J$ for the three different models: $J = 0.1$ eV (HM); $J = 0.12$ eV (4SCE with $K/J = 0.3$); $J = 0.15$ eV (EHM).

FIG. 3. (a) and (b): $M_r/\rho$ as a function of the inverse system size. (c) $\rho/\rho_{\text{Raman}}$ [24]. We show ED data for the HM (triangles), for the HM with 4SCE and $K/J = 0.3$ (circles), and in an EHM with $K/J = 0.34$, $J_2/J = 0.04$, and $J_3/J = 0.03$ (squares). The dashed line is a linear $1/N$ extrapolation for $N > 16$ in the HM. The dotted lines are guides to the eye traced parallel to the Heisenberg extrapolation. The experimental values are computed from the Sr$_2$CuO$_2$Cl$_2$ data [18,29]. The error bar reflects indeterminacies in background subtraction and, for $\rho_{\text{Raman}}$, the dependence on excitation energy [29].

The additional terms are spin exchange interactions for next-nearest neighbor sites ($J_{ij} = J_2$) and for next-next-nearest neighbor sites ($J_{ij} = J_3$) in Eq. (1). In order to be systematic we consider therefore an extended Heisenberg model with all magnetic interactions arising at fourth order.

To estimate the parameters we followed Ref. [10] and apply the perturbative expressions to an extended Hubbard model including second ($t_{2\text{nd}}/t_{1\text{st}} = 0.15$) and third ($t_{3\text{rd}}/t_{1\text{st}} = -0.12$) neighbor hoppings $t$. The Hubbard $U$ is taken as $8t_{1\text{st}}$. This approach has been shown to be consistent with MR, neutron-derived spin-wave velocity as well as angular-resolved-photoemission spectroscopy data [10]. From now on we refer to the resulting magnetic Hamiltonian as the extended Heisenberg model (EHM).

In Fig. 2 we show the line shape and in Fig. 3 we show the size dependence of the moments. We see that the effect of these terms also improves the agreement. Again we notice a strong sensitivity of the spectra to the underlying magnetic Hamiltonian.

A fine-tuning of the Hamiltonian parameters, which accurately reproduce the experiments, is not possible.
with the limited sizes available, due to the difficulty of a precise extrapolation of the cumulants to an infinite system. Clearly a sizable value of $K/J$ is needed and probably also a non-negligible $J_2/J$ and $J_3/J$. Our best estimate is the latter parameter set considered. In fact, a rough extrapolation from Fig. 3, assuming a similar scaling as for the HM, gives a result quite close to the experimental data. We mention that by setting $K = 0$ in the EHM we were not able to obtain an acceptable fit to the experimental data with reasonable values of $J_2/J$ and $J_3/J$. Even for the EHM Fig. 2 does not show perfect agreement between theory and experiment but the moment analysis shows that this can be ascribed to a finite size effect.

The energy scale can be fixed by matching the first moment with the experimental first moment. In the EHM we find $J \sim 0.19$ eV for Sr$_2$CuO$_2$Cl$_2$ in good agreement with the value found in Ref. [10] using other spectroscopic data. As an alternative procedure one can adjust the position of the first peak as done in Fig. 2. This gives a somewhat smaller value probably due to finite size effects. These values of $J$ are not in contradiction with the smaller value of $J$ usually quoted in cuprates ($J \sim 0.1-0.13$ eV): A spin-wave theory computation shows that, at low energies, the effect of the extended terms in the Hamiltonian is to renormalize the effective $J$ to lower values [7].

We also computed the staggered magnetic moment in the EHM. We get a staggered moment roughly 7% larger than in the HM (see also Refs. [9,26]). This may be important in view of the disagreement found for this quantity between theory and experiment [26,27].

In conclusion we have presented a computation of the IR absorption spectra due to magnetic excitations in undoped cuprates. We have shown that these experiments are very sensitive to the underlying magnetic Hamiltonian. We find that the usual model used to describe the spin dynamics in cuprates, namely, a HM with nearest neighbor exchange, cannot explain the experimental data. Instead an EHM with further neighbor interactions and with a 4SCE term is in good agreement with the data. To the best of our knowledge this provides the first quantitative explanation of the puzzling sidebands in the spectrum. In addition, the same model has been found to agree with other spectroscopic data [10]. Further theoretical work is needed to explore the consequences of the 4SCE term in the doped phase.

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[17] We argue that the interpretation of the MR line shape is not conclusive because: (i) The scattering mechanism itself is under discussion since resonant effects are important [28]. In fact, a strong dependence of the line shape on the excitation energy has been reported [29] and one can see that both anomalies change substantially from one experimental group to the other. (ii) Alternative explanations for the width of the two-magnon peak exist without invoking the 4SCE term [11].
[24] $\rho_{\text{Raman}}$ is for an infinite cluster. This extrapolation will be discussed elsewhere.