Higgs Mode and Magnon Interactions in 2D Quantum Antiferromagnets from Raman Scattering

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We present a theory for Raman scattering on 2D quantum antiferromagnets. The microscopic Fleury-Loudon Hamiltonian is expressed in terms of an effective $O(3)$ - model. Well within the Néel ordered phase, the Raman spectrum contains a two-magnon and a two-Higgs contribution, which are calculated diagrammatically. Due to the momentum dependence of the Raman vertex in the relevant $B_{1g} + E_{2g}$ symmetry, the contribution from the Higgs mode is strongly suppressed. It shows up as separate peak in the spectrum only for an intermediate value of the Higgs mass. The dominant contribution to the spectrum arises from two-magnon excitations. They lead to a broad, asymmetric peak which is a result of magnon-magnon interactions mediated by the Higgs mode. The associated vertex function is determined from a numerical solution of the Bethe-Salpeter equation. The resulting spectrum, which has the antiferromagnetic exchange coupling as a single parameter, turns out to be in very good agreement with Raman scattering data on undoped cuprates.

I. INTRODUCTION

The recent observation of a scalar Higgs particle as the final missing ingredient of the standard model in particle physics has led to a resurgence of interest in the question whether a well defined excitation associated with an amplitude mode is present and observable in condensed matter systems with a broken continuous symmetry. A case in question are neutral superfluids, in which a global $U(1)$ symmetry is broken as the simplest example of continuous symmetry breaking. The dynamics of the superfluid order parameter, however, is generically of the Gross-Pitaevskii type, i.e. first order in time. Despite the Mexican hat structure of the effective potential, there is thus only a Goldstone but no Higgs mode. Indeed, a necessary condition for a Higgs mode in the non-relativistic context of condensed matter systems is an emergent Lorentz invariance. This appears, for instance, in the superfluid phase of ultracold Bosons in an optical lattice at integer filling, where the particle-hole symmetry close to the transition to a Mott -insulating phase gives rise to an effectively Lorentz invariant action. In this regime, a Higgs mode has indeed been observed from the absorbed energy in response to shaking the optical lattice. The associated Higgs mode energy vanishes in a continuous manner at the transition to a Mott insulator, essentially like a mirror image of the gapped particle-hole excitations in the incompressible phase. A Higgs mode has also been observed in neutron scattering from the insulating magnet ThCulCl$_3$, which exhibits a transition from a dimer spin liquid to a Néel ordered phase. In both cases, the evolution of the Higgs mode can be followed into the regime where the order parameter vanishes near a quantum critical point. In the context of superconductors at very low temperatures, where the dynamics of the order parameter is again second order in time, a Higgs mode has been observed in a pioneering experiment by Sooryakumar and Klein via Raman scattering. The possibility to detect the amplitude mode associated with oscillations of the superconducting gap via Raman scattering requires, however, a rather special situation with a coexisting charge density wave, as appears in NbSe$_2$. Indeed, in this case, the superconducting order parameter directly couples to light because the modulation of the charge density wave by the electromagnetic field in the $A_{1g}$ symmetry of the Raman response leads to a periodic modulation of the density of states at the Fermi surface and thus of the superconducting gap. This interpretation of the results obtained by Sooryakumar and Klein has been confirmed in detail by a recent experiment.

In our present paper, we analyze the possibility to observe a Higgs mode in quantum spin models whose ground state exhibits antiferromagnetic order in two dimensions via Raman scattering. This problem is of interest for at least two different reasons: First of all, there are experimental data on clean samples of undoped cuprates for which no theoretical model and understanding seems to have been given so far. Second, due to its relevance in the parent compounds of high temperature superconductors, quantum antiferromagnets in two dimensions are among the most intensively studied examples of a broken continuous symmetry in condensed matter. Somewhat surprisingly, the question about possible signatures of a Higgs mode in this context appears to have hardly been studied so far. This question is of interest also from a quite general point of view because the issue of a well defined Higgs excitation is nontrivial in two dimensions (2D). Indeed, in this case, the phase space for the decay of a Higgs mode into two Goldstone modes is very large. As a result, the Higgs spectrum at zero wave vector extends down to zero energy, diverging with a universal power law $\sim 1/\omega$. To leading order, the associated low energy singularity is due to two-magnon processes, as observed in neutron scattering from the Néel state of undoped LaCu$_2$CuO$_4$. The presence of a large weight in the spectrum of the amplitude mode at low energies suggests that the Higgs mode is overdamped in 2D. As emphasized by Podolsky, Auerbach and Arovas, this is not the case in general, however. In particular, response functions which - unlike neutron scattering - cou-
ple to the square of the order parameter are expected to show a clear Higgs peak even in two dimensions because for them the low frequency response is suppressed by a factor $\omega^3$. Precisely this type of response is measured in the lattice shaking experiment near the superfluid to Mott insulator transition performed by Endres et al. As a result, a sharp peak due to a Higgs excitation appears even in 2D, as supported by a detailed theoretical analysis of this problem, both numerically \[^{3,16,17}\] and analytically \[^{18}\]. Moreover, the Higgs mode remains well defined even near the quantum critical point because its energy and width vanish with the same power in the distance from the critical point \[^{3,16-19}\].

Regarding the Néel ordered state of 2D quantum antiferromagnets, Raman scattering seems to be ideally suited to observe the amplitude mode since it couples to the square of the order parameter \[^{15}\]. Experimentally, however, no indications for a Higgs mode are found \[^{10}\]. Instead, there is a broad, asymmetric peak which appears to be due to a strongly renormalized two-magnon excitation. As will be shown below, this observation can be explained both in qualitative and quantitative terms within a rather simple O(3) \(^{-}\) model of the Néel ordered phase. In particular, the absence of a Higgs mode in Raman scattering is a consequence of the fact that the leading order contributions from the amplitude mode of the order parameter vanish because of the nontrivial Raman vertex in the relevant B_{1g} symmetry. It is only for an intermediate value of the mass term in the underlying O(3) \(^{-}\) model where a small additional peak due to the Higgs mode appears above the two-magnon continuum. Such a feature is observed in the Raman spectrum of La$_2$CuO$_4$ \[^{11}\]. As will be discussed below, however it is unclear whether this feature can be associated with the Higgs mode of the Néel ordered state. In the dominant two-magnon response, the Higgs mode shows up only indirectly by modifying the magnon-magnon interactions, which crucially affect the detailed form of the Raman spectrum. Remarkably, the experimentally observed spectra for both La$_2$CuO$_4$ and YBa$_2$Cu$_3$O$_6$ are in very good agreement with the theory, basically without any adjustable parameter.

II. RAMAN SPECTRUM

A. Effective light scattering operator

The differential cross section for Raman scattering is given by \[^{20}\]

$$\frac{d^2\sigma}{d\Omega d\omega} = \hbar r_0^2 \frac{\omega_i}{\omega_f} R(i \rightarrow f)$$

(1)

where $r_0 = \frac{e^2}{m_e c^2}$ is the Thompson radius and $R(i \rightarrow f)$ is the transition rate from the initial photon state $|k_i, e_i\rangle$ to the final state $|k_f, e_f\rangle$. The rate is determined by Fermi’s golden rule \[^{20,21}\]

$$R(i \rightarrow f) = \frac{1}{\pi} \sum_{I,F} e^{-\beta E_I}|M_{F,I}|^2 \delta(E_F - E_I - \hbar \omega)$$

(2)

where $|I\rangle$, $|F\rangle$ are the initial, final state of the sample, $\hat{M}$ is the effective light scattering operator and $\omega = \omega_i - \omega_f$ is the frequency transfer. For the specific example of the Mott-insulating, antiferromagnetic phase of undoped cuprates, an appropriate microscopic Hamiltonian is the nearest neighbor Heisenberg model on a square lattice which arises from a superexchange mechanism \[^{22}\]. The associated effective light scattering operator is then given by the Fleury-Loudon Hamiltonian \[^{21,23,24}\] $\hat{H}_{FL}$ in leading order of a moment expansion. Performing the common symmetry decomposition of the Raman scattering operator \[^{20}\] it turns out that only the $B_{1g}$-mode is Raman active because the operators of the other symmetry modes commute with the Heisenberg Hamiltonian. Restricting the interaction to this mode, the reduced effective Hamiltonian is given by

$$\hat{H}_{FL} = 2B P(e_i, e_f) \sum_j \left( S_{x_j} \cdot S_{x_j+k} - S_{x_j+k} \cdot S_{x_j+y} \right)$$

(3)

with $P(e_i, e_f) = e_i^x e_f^x - e_i^y e_f^y$ and $B \sim t^2/(U - \hbar \omega_f)$. As has been shown by Shastry and Shraiman \[^{21}\], there is also a response in other symmetry modes associated with ring exchanges on a plaquette or the fluctuations of a chiral spin operator \[^{25}\]. They require an extension of the microscopic Hamiltonian beyond the Heisenberg model with nearest neighbor exchange only and thus lead to corrections to the simple form of the Fleury-Loudon Hamiltonian \(^{3}\). More generally, as will be discussed briefly in section II.E below, at higher frequencies even an extended Heisenberg model including e.g. four-spin interactions may not be adequate to describe the Raman response and one has to go back to the underlying Hubbard model to account for all possible excitations. In our present work, we do not consider such extensions. In fact, as will be shown below, already the leading order Hamiltonian \(^{3}\), which describes 2D quantum antiferromagnets with a well defined Néel order, properly accounts for the essential features of the Raman spectrum. In the following, therefore, any contributions beyond this leading order term are neglected. Apart from microscopic prefactors, the transition rate in Eq. (2) can then be written as the Fourier transform of the van Hove function $S(t)$ of $\hat{H}_{FL}$

$$R(i \rightarrow f) = \frac{e^2}{\hbar^3 c^2} g(k_i)g(k_f) \int_{-\infty}^{\infty} dt e^{i\omega t} \langle \hat{H}_{FL}(t)\hat{H}_{FL}(0) \rangle_T$$

(4)

The resulting Raman response function $S(\omega)$ is connected to an associated spectral function $\chi_{Raman}(\omega)$ by

$$S(\omega) = -\frac{1}{\pi} \text{Im} \chi_{Raman}(\omega)$$

where $\text{Im} \chi_{Raman}(\omega)$ is the imaginary part of $\chi_{Raman}(\omega)$.
the fluctuation-dissipation theorem. At zero temperature, this takes the simple form

$$S(\omega) = 2\hbar \chi_{\text{Raman}}(\omega) \theta(\omega) \equiv 2\hbar \text{Im} \chi_{\text{Raman}}(\omega + i0) \theta(\omega)$$

(5)

In practice, the spectral function will be determined from the imaginary time ordered correlation function

$$\chi_{\text{Raman}}(\tau) = \frac{1}{\hbar} \left< T_\tau \left[ \hat{H}_{\text{FL}}(\tau) \hat{H}_{\text{FL}}(0) \right] \right>$$

(6)

by analytic continuation to real frequencies.

### B. Field theoretic formalism

In order to disentangle the contributions from magnons and the Higgs mode to the Raman spectrum, it is both necessary and convenient to replace the spin-operators of the microscopic Heisenberg model by a coarse grained description in which the relevant excitations appear more directly. Such an effective description is provided by the $O(3)$ nonlinear-$\sigma$-model (NL$\sigma$M), which is the continuum field theory of the antiferromagnetic Heisenberg model as used by Chakravarty, Halperin and Nelson. The associated effective action can be written in an apparently relativistic invariant form

$$S[\mathbf{n}] = \frac{1}{2g} \int d^3x \left( \partial_\mu \mathbf{n} \right)^2, \quad g = \frac{\hbar c_s}{\rho_s}$$

(7)

with a three-vector $(x^\mu) = (c_s^\alpha, r, \tau, x)$ and spin wave velocity $c_s$. Apart from $c_s$ and the momentum cutoff $\Lambda$, the dimensionless coupling constant $\hat{g} = g\Lambda$ of the nonlinear-$\sigma$-model contains the spin stiffness $\rho_s$, which is of the order of the exchange coupling constant of the underlying Heisenberg model. The slowly varying order parameter field $\mathbf{n}(x)$ is a three component field which describes deviations from perfect Néel order. It is normalized by $\mathbf{n}^2 = 1$. On the microscopic level, the physical meaning of the field $\mathbf{n}$ becomes evident by the mapping

$$\frac{\hbar}{S} \mathbf{n}(x_j) \rightarrow (-1)^{x_3(x_j)} \mathbf{n}(x_j) \sqrt{1 - a^4 \mathbf{L}(x_j)^2 + a^2 \mathbf{L}(x_j)}$$

(8)

between spin operators and bosonic fields originally due to Haldane. Here $a$ is the lattice constant and the angular momentum field $\mathbf{L}$ is constrained by $\mathbf{n} \cdot \mathbf{L} = 0$, $a^2 \mathbf{L} \ll 1$. Using Eq. (8) to express the spin operators in the Fleury-Loudon Hamiltonian (3) in terms of the order parameter field, it turns out that the Raman spectrum requires to determine the connected correlation function of the following operator in the NL$\sigma$M

$$O(\tau) = 2B \hbar^2 S^2 P(e_i, e_f) \int d^2x \, \sigma_{ij} \partial_\tau \mathbf{n}(x, \tau) \cdot \partial_\tau \mathbf{n}(x, \tau)$$

$$= 4B \hbar^2 S^2 P(e_i, e_f) \int \frac{d^2k}{(2\pi)^2} \gamma(k) |\mathbf{n}(k, \tau)|^2$$

(9)

Here $\gamma(k) = \frac{1}{2} (k_x^2 - k_y^2)$ is the continuum form of the $B_1g$-symmetry factor. Unfortunately, the combination of nontrivial vertices in the NL$\sigma$M which appear beyond a simple Gaussian approximation due to the constraint $\mathbf{n}^2 = 1$ and the complicated form of the operator (9) make it very hard to calculate the Raman spectrum directly within the NL$\sigma$M. In particular, it is rather difficult to disentangle the contributions which arise from magnons or the Higgs excitation in the standard decomposition $\mathbf{n} = (\mathbf{\pi}, \sqrt{1 - \mathbf{\pi}^2})$ which only involves the two-component field $\mathbf{\pi}$ associated with the magnons. It is therefore more convenient to use a soft-spin description of the order parameter within a linear $O(3)$-$\sigma$-model. Its effective action

$$S[\Phi] = \frac{1}{2g} \int d^3x \left[ (\partial_\mu \Phi)^2 + m_0^2 \left( |\Phi|^2 - 3 \right)^2 \right]$$

(10)

contains the bare mass $m_0$ of the amplitude mode as an additional parameter. Fortunately, as will be shown below, the Raman spectrum in the relevant limit of $\hat{g} \ll 1$ is hardly sensitive to the precise value of $m_0$. It is fixed essentially by the spin stiffness or the equivalent Heisenberg exchange energy $J$, which is known rather accurately from neutron scattering data. As a result, the description of 2D quantum antiferromagnets in terms of a linear $O(3)$-$\sigma$-model is convenient not only in terms of its simpler diagrammatics but also does not - in effect - introduce an additional adjustable parameter. An intuitive argument for why it is possible to replace the fixed length NL$\sigma$M with a soft-spin model relies on viewing the linear $\sigma$-model as an effective low-energy theory of the NL$\sigma$M, where short wave-length fluctuations have been integrated out. The Neel-field $\mathbf{n}(x)$ is therefore averaged over some domain in real space and the resulting averaged field $\Phi$ is no longer constrained to have a fixed length. Instead, it is subject to a potential $V(\Phi)$ which exhibits a minimum for $\Phi^2 = 1$ (note that in Eq. (10), we have rescaled the field by a factor $\sqrt{3}$, so that its vacuum expectation value (VEV) obeys $\langle |\Phi|^2 \rangle = \sqrt{3}$). The magnitude of the fluctuations around the minimum is controlled by the dimensionless mass parameter $\hat{m}_0 = m_0 / \Lambda$, which is expected to be large deep in the Néel ordered phase. The dynamics of the averaged field is fixed by the condition that the magnon dispersion $\omega_k = c_s |k|$ is linear in momentum, giving rise to an effectively Lorentz invariant action.

In order to calculate the Raman spectrum within the linear $\sigma$-model (10), we use the standard parametrization for the field $\Phi$ in the symmetry broken phase:

$$\Phi = \left( \mathbf{\pi}, r \sqrt{3} + \sigma \right).$$

(11)

The parameter $r = r(g, \Lambda)$ incorporates the renormalization of the VEV by quantum fluctuations and has to be determined from the condition $\langle \sigma \rangle = 0$. The components of $\mathbf{\pi}$ are the two massless antiferromagnetic magnons, while $\sigma$ denotes the massive amplitude/Higgs mode. Expressed in terms of the relativistic three-momentum $k = (c_s r, \sqrt{3} + \sigma)$ the spectral function will be determined from the imaginary time ordered correlation function

$$\chi_{\text{Raman}}(\tau) = \frac{1}{\hbar} \left< T_\tau \left[ \hat{H}_{\text{FL}}(\tau) \hat{H}_{\text{FL}}(0) \right] \right>$$

(6)
\( (\omega/c_s, k) \), the free propagators for the fields are

\[
G^0_{\pi\pi}(k) = \langle \pi_i(k)\pi_j(-k) \rangle = \delta_{ij}G^0_{\pi\pi}(k) = \delta_{ij}\frac{g}{k^2}
\]

\[
G^0_{\sigma\sigma}(k) = \langle \sigma(k)\sigma(-k) \rangle = \frac{g}{k^2 + m_0^2}
\]

These propagators will change with increasing strength \( g \) of the quantum fluctuations. In particular, the exact propagator

\[
G_{\sigma\sigma}(k) = \frac{g}{k^2 + m_0^2} + \frac{g^2}{(k^2 + m_0^2)^2} \frac{m_0^4}{24|\vec{k}|} + \ldots
\]

of the Higgs mode acquires a contribution \( \sim 1/|\vec{k}| \) due to the decay into Goldstone modes which appears at order \( g^2 \). Instead of a simple pole, it therefore exhibits a branch cut which starts at the threshold \( \omega_k = c_s|\vec{k}| \) for the excitation of magnons. By contrast, the Goldstone theorem enforces that the magnons are massless, i.e. the structure \( G_{\pi\pi}(k) = Zg/k^2 \) remains exact with a finite, renormalized coupling constant \( Zg \) throughout the Néel ordered phase. Inserting eq. (11) into eq. (9) we obtain

\[
O(\tau) = O_{\text{Magnon}}(\tau) + O_{\text{Higgs}}(\tau)
\]

\[
O_{\text{Magnon}}(\tau) \propto \int d^2x \left[ (\partial_x \pi)^2 - (\partial_y \pi)^2 \right]
\]

\[
O_{\text{Higgs}}(\tau) \propto \int d^2x \left[ (\partial_x \sigma)^2 - (\partial_y \sigma)^2 \right]
\]

The operator to which light couples in the Raman response therefore involves the square of gradients of the magnon and Higgs field. As a result, the response always involves at least two magnon or Higgs excitations. Taken together, the full Raman susceptibility is the sum of three different contributions

\[
\chi_{\text{Raman}}(\omega) = \chi_M(\omega) + \chi_H(\omega) + \chi_{\text{Int}}(\omega)
\]

which involve a two-magnon susceptibility \( \chi_M(\omega) \) and a two-Higgs susceptibility \( \chi_H(\omega) \). They are determined by the correlation functions of two \( O_{\text{Magnon}} \) and two \( O_{\text{Higgs}} \) operators, respectively. In addition, an interference susceptibility \( \chi_{\text{Int}}(\omega) \) arises which consists of the two mixed correlation functions. A similar structure has in fact been found by Canali and Girvin, who have calculated the Raman spectrum of undoped cuprates by means of a Dyson-Maleev transformation of the underlying antiferromagnetic Heisenberg model. Within this formulation, there is no clear distinction between the magnon and Higgs contributions to the spectrum, with the latter appearing as part of a four-magnon term.

To leading order in the coupling \( g \), only the two-magnon and two-Higgs susceptibilities are nonzero while the interference susceptibility contributes only at order \( g^3 \). The corresponding Feynman diagrams in FIG. 1 are just a two-magnon, respectively a two-Higgs bubble dressed with two Raman symmetry factors \( \gamma(\vec{k}) \). Dropping the polarization factor \( P(e_\pi, e_\sigma) \), the two-magnon response to lowest order in \( g \) is given by

\[
\chi_M''(\omega) = \frac{g^2B^2h^3S^4}{12c_s} \omega^3 \theta(2c_s\Lambda - \omega) + O(g^3).
\]

The spectrum is a pure power law \( \sim \omega^3 \) with a sharp cutoff at twice the zone-boundary energy of a magnon. If we assume, that the Heisenberg model underlying the continuum theory contains only a nearest-neighbor coupling \( J \), this corresponds to \( 2c_s\Lambda = 2\pi h J \). The peak at the upper cutoff in the spectrum of (19) can then be understood to arise from processes in which the incoming photon flips the spins on two neighboring sites, which costs the energy \( 6h^2 J \) in a square lattice.

The two-Higgs bubble gives rise to a threshold at \( 2m_0 \). To leading order in \( g \) the associated Higgs contribution to the spectrum is given by

\[
\chi_H''(\omega) = \frac{g^2B^2h^3S^4}{12c_s} \left[ \left( \frac{\omega}{2c_s} \right)^2 - c_s^2 m_0^2 \right] \theta(\omega - 2c_s m_0)
\]

\[
\times \theta \left( 2c_s \sqrt{\Lambda^2 + m_0^2 - \omega} \right) + O(g^3).
\]

It shows the expected threshold at twice the Higgs mass and peaks at the maximum energy of two Higgs excitations. Note that in contrast to neutron scattering, the Raman spectrum does not contain the longitudinal susceptibility \( \chi_{\sigma\sigma} \sim \langle \sigma \sigma \rangle \). As a result, no sharp peak due to the Higgs mode appears even at leading order. In the following section, we will show that the sharp features in both the two-magnon and the two-Higgs response, which depend quite sensitively on the precise form of the momentum cutoff and the specific value of the Higgs mass \( m_0 \), are completely washed out in a calculation which sums up the next-to-leading order diagrams in a consistent fashion.
The next-to-leading order diagrams, apart from self-energy insertions, for the two-magnon, two-Higgs and interference susceptibility are shown in figures FIG. 2, FIG. 3 and FIG. 4 respectively. To see whether these diagrams may lead to sharp features associated with the Higgs mode, we start with a qualitative discussion based on symmetries alone. One immediately notices, that the loop integrals in FIG. 2(a) decouple. Since \( q = (\omega,0) \), these diagrams therefore give no contribution to the Raman response because of the antisymmetry of the \( B_{1g} \)-symmetry factor \( \tilde{\gamma}(k) \) under the exchange \( k_x \leftrightarrow k_y \). Especially the vanishing of diagram FIG. 2(b) is important to notice, as it would lead to a peak at the Higgs mass, due to the pole in the Higgs propagator. The only non-vanishing diagram for the two-magnon susceptibility is FIG. 2(c), in which the Higgs line is independent of the frequency transfer. As a result, no sharp peak at the Higgs mass arises.

For the two-Higgs and the interference susceptibilities the situation is quite similar. Indeed, the diagrams (a) and (b) in FIG. 3 and FIG. 4 vanish due to the antisymmetry of the \( B_{1g} \)-symmetry factor under the exchange \( k_x \leftrightarrow k_y \). Concerning the interference contribution, it turns out, that up to order \( g^3 \) it gives a negligible contribution to the full Raman response. Thus, in the following, we only consider the two-magnon and two-Higgs contributions.

### Beyond leading order and Bethe-Salpeter equation

To determine the Raman spectrum, we do not need the full vertex function. Instead, it is sufficient to know

\[
\chi_M(\omega) = 2 \int \frac{d^3 k}{(2\pi)^3} \int \frac{d^3 k'}{(2\pi)^3} \tilde{\gamma}(k) \tilde{\gamma}(k') G_{\pi\pi}(k, \Omega + \omega) G_{\pi\pi}(k', \Omega) \Gamma_{\pi}(k, k', \Omega, \Omega', \omega). \tag{21}
\]

Here, we have introduced the exact two-magnon Raman vertex function \( \Gamma_{\pi}(k, k', \Omega, \Omega', \omega) \). As is depicted diagrammatically in FIG. 6, the vertex function obeys a Bethe-Salpeter equation

\[
\Gamma_{\pi}(k, k', \Omega, \Omega', \omega) = 2(2\pi)^3 \delta(k - k') + \int \frac{d^3 k''}{(2\pi)^3} \mathcal{V}_{\pi}(k, k'', \omega) G_{\pi\pi}(k'', \Omega'' + \omega) G_{\pi\pi}(k'', \Omega'') \Gamma_{\pi}(k'', k', \Omega'', \Omega', \omega) \tag{22}
\]

in which the magnon propagators

\[
G_{\pi\pi}(p) = \frac{g}{p^2 - g\Sigma_{\pi\pi}(p)} \tag{23}
\]

are understood to be the full ones. They thus include all self-energy corrections, i.e. \( \Sigma_{\pi\pi} \) is the exact magnon self-energy. Moreover, the kernel in Eq. (22) contains the full irreducible two-magnon interaction \( \mathcal{V}_{\pi}(k, k'', \omega) \).

To determine the Raman spectrum, we do not need the full vertex function. Instead, it is sufficient to know

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FIG. 2. Next-to-leading order diagrams apart from self-energy insertions for the 2-magnon susceptibility \( \chi_M \)

FIG. 3. Next-to-leading order diagrams apart from self-energy insertions for the 2-Higgs susceptibility \( \chi_H \)

FIG. 4. Next-to-leading order diagrams for the interference susceptibility \( \chi_{\text{int}} \)
The two-magnon interaction $V_\pi(k, k'', \omega)$ is given by

$$V_\pi(k, k'', \omega) = \frac{m_0^2}{2\sqrt{3}g} G^0_{\sigma\sigma}(k - k'') \Gamma_\pi(k', \Omega', \omega)$$

Thus, to lowest order, the interaction of magnons which is relevant for the $B_{1g}$ symmetry is mediated by the Higgs mode. Inserting Eq. (27) into Eq. (26), we obtain the ladder Bethe-Salpeter equation, which corresponds to a consistent next-to-leading order approximation in the irreducible magnon-magnon interaction. Regarding the magnon propagator, we use the result $G^0_{\pi\pi}$ to zeroth order in $g$. Apart from a numerical factor $g \to Z_g$, this is exact at small $k$ due to the Goldstone theorem. The final form of the Bethe-Salpeter equation in ladder approximation is therefore given by

$$\Gamma_\pi^{\text{Ladder}}(k, \Omega, \omega) = 2\tilde{\gamma}(k) + \frac{m_0^2}{3g^2} \int \frac{d^3k''}{(2\pi)^3} G^0_{\sigma\sigma}(k - k'') \times G^0_{\pi\pi}(k'', \Omega'') \Gamma_\pi^{\text{Ladder}}(k'', \Omega'', \omega) \times \Gamma_\pi^{\text{Ladder}}(k', \Omega', \omega) \times \Gamma_\pi^{\text{Ladder}}(k', \Omega', \omega) \times \Gamma_\pi^{\text{Ladder}}(k', \Omega', \omega). \quad (28)$$

This is still too complicated to be solved analytically, however it is amenable to an efficient numerical solution. Before, doing so, however, a similar equation will be derived for the two-Higgs susceptibility.

To derive the integral equations for the two-Higgs susceptibility is completely analogous to the case of the two-Magnon susceptibility, one just needs to replace the magnon propagators by Higgs propagators. Hence we will just write down the resulting equations, containing already the reduced two-Higgs Raman vertex function $\Gamma_\sigma(k, \Omega, \omega)$:

\[ \Gamma_\sigma(k, \Omega, \omega) = 2\tilde{\gamma}(k) + \int \frac{d^3k''}{(2\pi)^3} V_\sigma(k, k'', \omega) G_{\sigma\sigma}(k'', \Omega'') \times G_{\pi\pi}(k'', \Omega'' + \omega) \Gamma_\sigma(k'', \Omega'', \omega) \times \Gamma_\sigma(k'', \Omega'', \omega) \times \Gamma_\sigma(k'', \Omega'', \omega). \quad (29) \]

The leading diagrams for the full two-Higgs interaction $V_\sigma(k, k'', \omega)$ are depicted in FIG. 7. Truncating the irreducible two-Higgs interaction at lowest order in $g$, which amounts to a ladder approximation. Properly taking into account combinatorial factors we find

\[ \Gamma_\sigma^{\text{Ladder}}(k, \Omega, \omega) = 2\tilde{\gamma}(k) + \frac{m_0^2}{3g^2} \int \frac{d^3k''}{(2\pi)^3} G^0_{\sigma\sigma}(k - k'') \times G^0_{\pi\pi}(k'', \Omega'') \Gamma_\sigma^{\text{Ladder}}(k'', \Omega'', \omega) \times \Gamma_\sigma^{\text{Ladder}}(k', \Omega', \omega) \times \Gamma_\sigma^{\text{Ladder}}(k', \Omega', \omega). \quad (28) \]
order in $g$, we obtain

$$V_\sigma(k, k'', \omega) = 2 \cdot (2 \cdot 3)^2 \cdot \frac{1}{2} \left( \frac{m_0^2}{2\sqrt{dg}} \right)^2 G^0_{\sigma\sigma}(k - k'')$$

(31)

Note that this has precisely the same form as the irreducible two-magnon interaction, differing only in the combinatorial and coupling prefactors. Inserting eq. (31) into Eq. (30) we obtain the Bethe-Salpeter equation for the reduced two-Higgs Raman vertex function

$$\Gamma^\text{Ladder}_\sigma(k, \Omega, \omega) = 2\tilde{\gamma}(k) + \frac{3m_0^2}{g^2} \int d^3k'' \frac{G^0_{\sigma\sigma}(k-k'')}{(2\pi)^3} G^0_{\sigma\sigma}(k''-\Omega')$$

(32)

which again needs to be solved numerically.

D. Numerical solution of the Bethe-Salpeter equation and Raman spectra

For the numerical solution of the two independent Bethe-Salpeter equations (28) and (32), it is convenient to first write them in dimensionless form. This is achieved by introducing $k = \kappa/\Lambda$, $m_0 = m_0/\Lambda$, $\bar{g} = g\Lambda$, $\tilde{\chi}(\tilde{\omega}) = \chi(\omega)/\Lambda$ and $\bar{\Gamma}^\text{Ladder}(\kappa, \tilde{\omega}) = \Lambda^{-2}\Gamma^\text{Ladder}(k, k', \tilde{\omega})$. Note that the dimensionless coupling $\bar{g}$ is essentially given by the inverse of the spin quantum number $S$, $\bar{g} = 2\pi/S$ and that frequency is measured in units of $c_\sigma\Lambda \sim S$, which is important when comparing data at different dimensionless couplings.

The explicit numerical solution of the Bethe-Salpeter equation employs the standard approach to integral equations, namely discretizing the integrals and solving the resulting system of linear equations. For the dimensionless Brillouin-zone $[-1, 1]^2$, we use $20 \times 20$ points. Since an increase up to a grid size of $30 \times 30$ gave only very small corrections, we can assume that the results have converged to the continuum limit. While the frequency integration has no intrinsic microscopic cut-off, in practice the frequency integral is restricted to values below $4c_\sigma\Lambda$. Contributions beyond that do not affect our results. We have varied the number of sampling points for the frequency integral between 20 and 40. Similar to the situation for the momentum integration, the grid size had no influence on the results.

In FIG. 8, the two-magnon susceptibility $\tilde{\chi}_M(\tilde{\omega})$ obtained from the numerical solution of the Bethe-Salpeter equation is shown for three different values of the dimensionless coupling $\bar{g}$, corresponding to $S = 1/2, 1, 2$ and for an intermediate value of the Higgs mass $\tilde{m}_0 = 0.5$. To obtain the associated spectral function, which determines the Raman spectrum in real frequencies, an analytic continuation is required. Since our numerical data contain no noise, we use an Ansatz to fit the data which can then be continued in analytical form. Specifically, as a fit model, we use

$$\chi(\omega) = \frac{a_1 + a_2q^2}{1 + a_3q^2 + a_4q^4 + a_5q^6}$$

(33)

This Ansatz is motivated by our leading order results for the two-magnon susceptibility, which obeys $\chi_M(\omega) \sim \omega^{-2}$ for large $\omega$ and $\tilde{\chi}_M(\tilde{\omega}) \sim \tilde{\omega}^{-2}$ for small $\tilde{\omega}$. The fitted curves are also shown in FIG. 8. Apparently they reproduce the numerical results very well with a variance which is only $1.4 \times 10^{-6}$.

Based on the explicit form of the spectral function Eq. (33) in imaginary frequencies, the analytic continuation $q \to -i\tilde{\omega}$ is easily done. The resulting two-magnon spectral functions $\tilde{\chi}_M^{\text{re}}(\tilde{\omega})$ for $S = 1/2, 1, 2$ are shown in FIG. 9. For all values of $S$, a clear two-magnon peak appears. Taking into account the different frequency scales for different values of $S$, the peak is shifted towards lower frequencies for smaller spin. This behavior has already been observed by Canali and Girvin, which is essentially achieved for a
quantitative agreement with experiment.

In order to see to which extent the precise value of the bare Higgs mass \( \tilde{m}_0 \) influences our results for the two-magnon response, we have varied this parameter in the range \( 0.1 < \tilde{m}_0 < 0.9 \). Surprisingly, we find no detectable change in the two-magnon line shape. The physical reason for the fact that the two-magnon spectral function is essentially independent of the Higgs mass despite the fact that the magnon interaction is mediated by the amplitude mode, is the following: The inverse Higgs mass is the range of the interaction in position space while the interaction strength is controlled by the coupling \( g \). As both magnons, involved in the interaction process, are created by a single photon, they are initially on neighboring lattice sites. For \( \tilde{m}_0 < 1 \) the range of the Higgs-mediated interaction is larger than one lattice spacing, hence we do not expect a dependence of the 2-magnon peak on the Higgs mass.

As a second step, we turn to the results for the two-Higgs susceptibility where a similar dependence on the dimensionless coupling \( \tilde{H} \) appears as for the two-magnon response. We therefore only show the results for \( S = 1/2 \) which is relevant for cuprates. In FIG. 10 the imaginary time data for the two-Higgs susceptibility is shown for different Higgs masses. First of all one notices, that the absolute values of the two-Higgs susceptibility are smaller than the ones for the two-magnon susceptibility. As a result, the two-magnon peak will always dominate the Raman spectrum. From the leading order analysis, we again expect a \( \tilde{\omega}^{-2} \)-behavior for large frequencies. An Ansatz of the form used in Eq. (33) therefore again reproduces the numerical data rather well, with a variance of \( 1.8 \times 10^{-5} \). The resulting contribution \( \chi_H''(\tilde{\omega}) \) to the Raman spectrum in real frequencies is shown in FIG. 11.

As expected, the spectra are shifted towards higher frequencies with increasing values of the bare Higgs mass \( \tilde{m}_0 \). In particular, the spectral weight is rather small below \( 2\tilde{m}_0 \), reminiscent of the threshold behavior in leading order.

An exact result which determines the relative weight with which magnons or the Higgs mode appear in the Raman spectrum can be obtained by considering the ratio of the first moments of the two-magnon and two-Higgs susceptibilities \( \chi_M'' \) and \( \chi_H'' \). These moments can be expressed in terms of the exact propagators \( G_{\pi\pi}(k) \) and

\[
G_{\pi\pi}(k) = \frac{1}{\tilde{m}_0^2 - \tilde{\omega}^2 + \Sigma_{\pi}(k)}
\]

where \( \Sigma_{\pi}(k) \) is the self-energy of the \( \pi \)-mode. The results for the ratio of the first moments are shown in FIG. 12 and 13.

FIG. 9. 2-magnon spectral function \( \chi_{\text{2M}}''(\tilde{\omega}) \) for \( S = 1/2 \) (blue), \( S = 1 \) (black) and \( S = 2 \) (red).

FIG. 10. Numerical results for the 2-Higgs susceptibility as a function of imaginary frequency for \( \tilde{m}_0 = 0.9 \) (blue), \( \tilde{m}_0 = 0.65 \) (black) and \( \tilde{m}_0 = 0.25 \) (red). The solid curves are fits of eq. (33).

FIG. 11. 2-Higgs spectral function \( \chi_H''(\tilde{\omega}) \) for \( S = 1/2 \) and three different values of the Higgs-mass, \( \tilde{m}_0 = 0.9 \) (blue), \( \tilde{m}_0 = 0.65 \) (black) and \( \tilde{m}_0 = 0.25 \) (red).
A "shoulder" appears above the two-magnon peak as a mode \( \tilde{m}_0 \) magnon peak. Second, the regime of a heavy amplitude response is completely dominated by the two-tensor peak associated with an excitation of this mode. It is only in the intermediate regime \( 0.35 \lesssim \tilde{m}_0 \lesssim 0.7 \) where a "shoulder" appears above the two-magnon peak as a remnant of the sharp onset of a Higgs-mode in a leading order calculation. As will be discussed below, such a feature is present in the experimental Raman spectrum of La\(_2\)CuO\(_4\). Whether this feature can indeed be interpreted as a signature of an intermediate mass Higgs mode is, unfortunately, unclear at present.

E. Comparison with experiment

In the following, we compare our theoretical results for the Raman spectral function \( \chi''_\text{Raman} \) with experimental data for the undoped cuprate materials YBa\(_2\)CuO\(_6\) (Y-123)\(^{10}\) and La\(_2\)CuO\(_4\) (LCO)\(^{11}\). Despite the simplicity of our model, the experimental spectra turn out to agree rather well with theory. Specifically, the frequency scale \( c_s \Lambda \) of the underlying linear \( O(3) \) \( \sigma \)-model is related to the nearest-neighbor exchange coupling by \( hJ = \pi^{-1} c_s \Lambda^{32} \). For undoped curates, the relevant values for \( J \) are known from inelastic neutron scattering. The agreement between the optimal values for \( J \) obtained from our fit to the Raman data and the n-scattering results is therefore a crucial consistency test for both the model and our approximations involved in calculating the Raman spectrum. Since the absolute value of the measured intensity cannot be calculated reliably in theory and also depends on experimental details which are unknown (the data are plotted in arbitrary units), an overall prefactor \( A \) remains undetermined. The experimental data are therefore fitted with a function of the form \( A\chi''(\omega/(c_s \Lambda)) \) which only involves the frequency scale \( c_s \Lambda = \pi hJ \) as a physical parameter.
The experimental data for Y-123 together with the theoretical fit is shown in FIG. 14. Apparently, they are in very good agreement except for the range between 3500 cm$^{-1}$ and 4500 cm$^{-1}$, where more spectral weight appears than predicted by our theory. The additional feature is rather broad and thus is unlikely to be connected with an intermediate mass Higgs mode shown in Fig. 13. In fact, adjusting $\tilde{m}_0$ such that $\chi_0^g$ has spectral weight in this region leads to an increase well beyond the one observed in the Y-123 data. The most likely explanation of this shoulder is a triple resonance, as discussed by Morr and Chubukov$^{33}$, where the incoming light is in resonance with the charge gap of the Hubbard model. Such processes are clearly not captured by our model. Apart from that, however, the quite non-trivial form of the dominant two-magnon peak is reproduced quite well by our theory. In particular, the extracted value $J = 126$ meV for the exchange coupling is rather close to that obtained from inelastic neutron scattering, which is $120$ meV$^{34}$. In order to decide to which extent the unknown value of Higgs mass affects the comparison between theory and experiment, the data have been analyzed both in terms of the small and also the large Higgs mass scenario. As mentioned already in section II.D, it turns out that there is only a minor difference in the resulting curves. The experimental data therefore do not allow to infer a quantitative result for the Higgs mass.

For LCO, we show the data together with the theoretical fit in FIG. 15. Again, the two-magnon peak again is reproduced very well with a value $J = 149$ meV for the exchange coupling which is in excellent agreement with the INS result 143 meV$^{14}$. Contrary to the case of Y-123 and our theoretical result, the Raman spectrum of LCO exhibits a slight increase with frequency in the spectral range above 6000 cm$^{-1}$. This deviation is most likely due to the luminescence of defects in the LCO sample, while such an effect is absent in Y-123, because of a higher sample quality.

Concerning the pronounced shoulder above the two-magnon peak at about 4500 cm$^{-1}$, it turns out that this additional feature is reproduced quite well by choosing an intermediate value $\tilde{m}_0 = 0.595$ of the Higgs mass. The overall very good agreement between theory and experiment suggests, that the additional feature seen in the Raman spectrum of LCO is indeed associated with an amplitude mode of the underlying Néel state. Its absence in the case of Y-123 might be explained by the tendency that the Higgs mass is larger deep in the antiferromagnetic phase, i.e. for increasing $\rho_s \sim J$. The smaller exchange coupling compared to LCO and the presence of the additional feature due to the triple resonance may thus explain the absence of a separate peak due to the Higgs mode in Y-123.

Unfortunately, the arguments for the observation of a direct signature of the Higgs mode in the Raman spectra of LCO remain inconclusive at the present stage. First of all, it is not clear whether a difference in the exchange couplings $J$ of only about 15% is sufficient to change the Higgs mass sufficiently. Moreover, the additional feature in the $B_{1g}$ mode near 4500 cm$^{-1}$ in LCO also appears in the experimental data for the $A_{2g}$ mode, where no two-magnon response is present. By contrast, no feature is seen at any frequency in the $A_{2g}$ mode of Y-123$^{11}$. Assuming that it is only the Higgs mode which couples to $A_{2g}$, a finite response at a somewhat smaller frequency due to the expected smaller value of $m_0$ should also be present in Y-123.

To conclude, the simple linear $O(3)$ $\sigma$-model is able to explain quantitatively the major features of the Raman spectrum of the Néel state of undoped cuprates. The spectrum is dominated by a broad, asymmetric two-magnon peak. Its detailed form is a result of magnon-magnon interactions which are mediated by the Higgs mode. The relevant energy scale is fixed by the exchange coupling of the underlying Heisenberg model. The amplitude mode itself shows up only as a small additional feature on top of the two-magnon excitations and it only...
appears for an intermediate value of the dimensionless Higgs mass $\tilde{m}_0$. Whether the feature seen in LCO is due to an amplitude mode is unfortunately not clear at this point and requires further analysis.

III. CONCLUSION AND OPEN PROBLEMS

Our present study of possible signatures of a Higgs mode in 2D quantum antiferromagnets is certainly only a first step into a more detailed analysis of this problem. While it is remarkable that a description based on the linear $O(3)$ $\sigma$-model which only captures the low energy physics of antiferromagnets is able to account for the detailed form of the observed Raman spectrum of undoped cuprates, it is obvious that the issue of the Higgs mode in this context is still not resolved completely. Among the open questions which remain we mention two:

a) from a quite general point of view, what are suitable and experimentally accessible correlation functions where the Higgs mode in 2D quantum antiferromagnets can be seen in direct form and can also be studied into a regime where Néel order disappears into some more complex spin liquid state?

b) in the context of the specific model studied above, can one calculate the mass parameter $m_0$ from an underlying microscopic model and - in particular - go beyond the perturbative calculation of the Raman spectrum which is only reliable deep in the Néel phase?

The complexity of the correlation functions which enter the Raman spectrum makes both problems quite challenging. Still, already a better understanding of the microscopic values of $m_0$ should clarify the open question whether the observed additional feature seen in LCO above the two-magnon peak is due to an intermediate mass Higgs mode or has some different origin not connected to spin-excitations. Regarding the issue of more appropriate correlation functions where the Higgs mode might show up more directly than in a standard Raman experiment, an interesting option appears to be resonant inelastic X-ray scattering (RIXS)\(^5\). As has been show by van den Brink\(^6\), this technique allows to measure a momentum dependent four-spin correlation associated with the operator

$$\hat{O}_q = \sum_{k} J_k \hat{S}_{k-q} \cdot \hat{S}_{-k}$$

In contrast to Raman spectra, where a two-magnon response appears at zero external wave-vector, RIXS is thus sensitive to the overall dispersion of the spin excitations.

In the language of the NL$\sigma$M Eq. (36) corresponds to $\hat{O}_q \sim \int d^2x \epsilon_q \times (\partial_x n)^2$. In comparison to Eq. (9) one notices, that RIXS again couples to the square of field gradients, however in a symmetric form, without the antisymmetric tensor $\sigma_{ij}$, which is responsible for the vanishing of the leading diagrams associated with the Higgs mode. It might thus be possible that the Higgs mode in 2D quantum antiferromagnets is more directly observable in a RIXS type experiment, similar to the probe of a Higgs mode in the Bose-Hubbard model which is induced essentially by a modulation of the hopping amplitude \(^4\). Clearly, this requires further work.

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