Optical absorption of spin ladders

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We present a theory of phonon-assisted optical two-magnon absorption in two-leg spin-ladders. Based on the strong intra-rung-coupling limit we show that collective excitations of total spin \( S = 0, 1 \) and 2 exist outside of the two-magnon continuum. It is demonstrated that the singlet collective state has a clear signature in the optical spectrum.

\[ H = \sum_{l,\alpha} \left[ \frac{3}{4} S_l^\alpha S_{l+1}^\alpha + \lambda (S_{l+1}^\alpha S_l + S_l^\alpha S_{l+1}) - \frac{1}{2} \sum_{l,\alpha} S_l^\alpha S_{l+1}^\alpha = H_1 + H_2 + H_3 \right] \] (2)

where \( S_l = 0(1) \) for a rung singlet(triplet). The rung-spin eigenbasis is given by \( | s \rangle = (| \uparrow \downarrow \rangle - | \downarrow \uparrow \rangle)/\sqrt{2}, | t_x \rangle = -(| \uparrow \uparrow \rangle - | \downarrow \downarrow \rangle)/\sqrt{2}, | t_y \rangle = i(| \uparrow \uparrow \rangle + | \downarrow \downarrow \rangle)/\sqrt{2}, \) and \( | t_z \rangle = (| \uparrow \uparrow \rangle + | \downarrow \downarrow \rangle)/\sqrt{2} \) where the first(second) entry in the kets refers to a site on leg '1(2)' of the ladder. As usual \( S^\alpha | s \rangle = 0 \) and \( S^\alpha | t_\beta \rangle = i\varepsilon_{\alpha\beta\gamma} | t_\gamma \rangle \) with the Levi-Civita symbol \( \varepsilon_{\alpha\beta\gamma} \). The action of \( T^\alpha \) on the rung basis is given by \( T^\alpha | s \rangle = | t_\alpha \rangle \) and \( T^\alpha | t_\beta \rangle = \delta_{\alpha\beta} | s \rangle \).

For vanishing inter-rung coupling the ground state of (3) is a pure rung-singlet product-state \( | \rangle = \otimes | s \rangle \). The excited states are products of singlets and triplets \( | t_{m\alpha} \rangle = \otimes | t_\alpha \rangle \otimes | t_{m\alpha} \rangle \) with an excitation energy given by the number of triplets \( N | t_{m\alpha} \rangle \). At finite \( \lambda \) the action of the ladder-Hamiltonian on these product states can be read off easily from (3): (i) \( H_2 \) creates(_destroys) pairs of nearest-neighbor (NN) triplets of equal \( \alpha \)-index, (ii) gives a pair of NN sites in a relative state of one singlet and one triplet \( H_2 \) generates NN hopping of the triplet via exchange of the singlet and triplet, and finally (iii) \( H_2 \) induces NN interactions between triplets. Contributions (ii) and (iii) define a three-flavored hard-core Bose system with NN hopping as well as NN interactions. These contributions renormalize the spectrum to \( O(\lambda^2) \). Process (i) does not conserve the triplet number and changes the spectrum only to \( O(\lambda^4) \) at \( \lambda \ll 1 \). The essential physics of the bound states and optical absorption is independent of \( O(\lambda^2) \)-terms. They will be neglected hereafter. Based on this simplification it is convenient to split the Hamiltonian into a bare 'kinetic' part \( H_1 + H_2 \) and a two-particle interaction \( H_3 \). The bare one-triplet eigenstates \( | k\alpha \rangle \) of momentum \( k \) are

\[ | k\alpha \rangle = \frac{1}{\sqrt{N}} \sum_l e^{ikl} | t_{l\alpha} \rangle \] (3)

\[ \varepsilon_k = \langle k\alpha | H_1 + H_2 | k\alpha \rangle - \langle | H \rangle = 1 + \lambda \cos(k) \] (4)

where \( \langle k'\alpha | k\beta \rangle = \delta_{\alpha\beta} | k' \rangle | k \rangle \). This agrees with seminal work on spin-ladders [2]. Hereafter the ground state energy \( \langle | H \rangle \) is set to zero. The bare two-triplet eigenstates in the singlet sector must incorporate the symmetry \( | t_{l\alpha} t_{m\beta} \rangle = | t_{m\beta} t_{l\alpha} \rangle \) as well as the hardcore constraint \( | t_{l\alpha} | t_{l\beta} \rangle = 0 \)
where \( |Im, S\rangle = \sum_\alpha |t_{\alpha\alpha}t_{\alpha\alpha}|/\sqrt{3} \) refers to the singlet combination of two triplets in real space and \( \varepsilon_{kq} \) is the bare two-particle kinetic energy. \( k(2q) \) is the total (relative) momentum, \( \langle k'q'|kq \rangle = \delta_{kk'}\delta_{qq'} \) and \( k \in [-\pi, \pi], q \in [0, \pi]. \) (Anti)periodic boundary conditions apply to \( k(q) \) and \( k, q = -|k, -q|. \) A discussion of the remaining two-particle states in the triplet and quintet sector is deferred to appendix A.

After these preliminaries we are in a position to evaluate the optical absorption due to phonon-assisted two-magnon emission (PME). This absorption results from a magnetoelastic excitation induced by the incoming photon-field and has been considered first in the planar geometry of the CuO\(_2\) square-lattice of the HT\(_C\) materials \([2,13]\). Its microscopic justification can be applied with little change to the case of spin-ladders with corner-sharing Cu-O structure. For the effective coupling one obtains

\[
H_{LS} = -E(t) \sum_{l,\alpha,\mu} \left[ p_{l\mu}u_{\mu\alpha\alpha} + p_A(2u_{\mu\alpha\alpha} - u_{\alpha\mu\alpha}) \right] S^\alpha_{\mu l}S^\alpha_{\mu l+1} \tag{7}
\]

\( E(t) \) is the time-dependent electric field which is polarized along the ladder. \( u_{\mu\alpha\alpha}(l,R) \) denote displacement coordinates of the oxygen site on the \( \mu \)-th leg of the ladder and refer to the oxygen sites in cell \( O(\text{right}) = l + 1, \) \( L(\text{left}) = l - 1, \) and \( R(\text{right}) = l + 1. \) The effective charges \( p_l \) and \( p_A \) can be derived from an expansion up to first order in \( u_l \) and \( E(t) \) of the Cu-O-Cu superexchange coupling. Expressions for \( p_l \) and \( p_A \) can be found in \([2,14]\). For cuprates which are strongly covalent materials and for \( E(t) \) polarized parallel to the ladder, \( |p_A| \gg |p_l| \) is a reasonable approximation \([13]\). Although the specific form of \([\bar{1}]\) depends on the corner-sharing Cu-O structure it is conceivable that a similar PME is also possible in other spin-ladder compounds. Introducing the Fourier transformed phonon-operators

\[
u_{\mu\alpha} = (a_{\mu\alpha} + a_{-\mu\alpha}^\dagger)/\sqrt{2M\omega_k} \]

with the phonon dispersion \( \omega_k \) and the reduced O mass \( M \) and replacing \( S^\alpha_{\mu l} \) with the bond variables \( S^\alpha_l \) and \( T^\alpha_l \) we get

\[
H_{LS} = E(t) \frac{1}{\sqrt{2}} \sum_{k,\mu} \left[ a_{\mu\alpha} + a_{-\mu\alpha}^\dagger \right] P_k \tag{8}
\]

\[
P_k = -\frac{1}{4\sqrt{NM\omega_k}} \sum_{l,\alpha} e^{ikl} \left[ p_l + 2p_A(1 - \cos(k)) \right] T^\alpha_l T^\alpha_{l+1} \tag{9}
\]

where terms proportional to \( S^\alpha_l S^\alpha_{l+1} \) and \( S^\alpha_l T^\alpha_{l+1} \) have been dropped since they do not act on a pure singlet product-state. Photon momentum conservation has been neglected since the wave length of light is large compared to the lattice spacing.

\[
\varepsilon_{kq} = 2[1 + \lambda \cos(k/2) \cos(q)] \tag{6}
\]
where the bare two-particle resolvent \( G(k, z) \) is given by

\[
G(k, z) = \frac{1}{4\pi} \int_{-\pi}^{\pi} dq \frac{\sin^2(q)}{z - (2 + 2\lambda \cos(k/2) \cos(q))} = \frac{\text{sign}[\text{Re}(a)]\sqrt{a^2 - 1} - a}{4\lambda \cos(k/2)} \tag{15}
\]

with \( a = (2 - z)/(2\lambda \cos(k/2)) \). The zeros at \( \omega = E^S_k \) and momentum \( k \) of the denominator on the r.h.s. of (4) correspond to (anti)bound states in the two-triplet singlet sector. Since both, the boundaries of the two-particle continuum \( \omega \) as well as the solutions of (11) are functions of \( k \) only if energies \( \omega \) are rescaled in terms of the quantity \( (\omega - 2)/\lambda \), the k-space structure of the rescaled two-particle spectrum is independent of \( \lambda \) within our approximation. In particular, a singlet bound-state exists at all \( k \) in the Brillouin zone. At \( k = \pi \) its binding energy is largest with \( E^S_k = 2 - \lambda \) while at \( k = 0 \) the binding energy is zero. This is shown in fig. 2, which depicts the bound-state dispersion along with the two-particle continuum. Also shown are triplet \( (S = 1) \) and quintet \( (S = 2) \) (anti)bound-states, which are optically inactive. These states are discussed in appendix C. Recently, similar collective states have been predicted using bond-boson techniques [13,17].

From (13,14,15) we can obtain the absorption by numerical integration. The rescaled intensity \( \lambda I(\tilde{\omega})/p_A \) is a function of \( \tilde{\omega} = (\omega - 2)/\lambda \) and \( p_I/p_A \) only and does not depend on \( \lambda \). Figure 2 depicts the absorption spectrum for \( p_I = 0 \). We note that our choice of the frequency variable \( \tilde{\omega} \) implies that zero incoming photon energy corresponds to the point \( -2/\lambda \) on the x-axis of fig. 2. For any finite spin gap, i.e. \( \lambda < 1 \), this point is off the range plotted which refers only to the frequency window \( -2 \leq \tilde{\omega} \leq 2 \) in which absorption occurs. Figure 2 demonstrates that the bound state has a profound impact on the absorption spectrum which comprises almost completely of a structure due to the integrated density of states of the bound state. In particular there is a van-Hove singularity at \( \omega = 2 - \lambda \) which results from the maximum in the bound-state dispersion at \( k = \pi \). Only in the special case of \( p_I = -4p_A \) this van-Hove singularity disappears. The inset in fig. 2 focuses on the remaining spectral weight. It has a maximum at the center of the two-triplet continuum, however its weight is very small as compared to the contribution from the bound states. At finite values of \( p_I \) a step-like feature appears at the spin-gap and for \( p_I < 0 \) a dip occurs due to interference in the effective charge \( p_k \). At present we believe that such features are of academic interest only. Yet, for completeness a typical spectrum displaying the latter effects is shown in fig. 3.

The preceding discussion does not rule out an interpretation of van-Hove like structures in an experimentally determined absorption spectrum solely in terms the momentum integrated bare two-triplet continuum rather than in terms of bound states. However, taking this point of view and using that \( |p_I/p_A| << 1 \), which is most likely the case, the absorption spectrum would rather be symmetric with respect to the center of gravity of the measured spectrum. Finally we note, that in case of a finite dispersion of the phonon additional structures may appear which are not contained in our present analysis.

In conclusion we propose that singlet bound-states have a dramatic impact on the optical absorption of dimerized spin-ladder systems which allow for phonon-assisted multi-magnon absorption of the Lorenzana-Sawatzky type. We hope that our work may stimulate further experimental investigation of this issue.

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APPENDIX A: $S=1,2$ STATES

The triplet and quintet two-particle states in real space correspond to the combinations $|lm, \alpha\rangle = \sum_{\beta, \gamma} \varepsilon_{\alpha \beta \gamma} |t_{l\alpha} t_{m\beta} \rangle / \sqrt{2}$ and $|lm, \alpha\rangle = \sqrt{3/8} |t_{l\alpha} t_{m\beta} \rangle + |t_{l\beta} t_{m\alpha} \rangle - \delta_{\alpha \beta}(2/3) \sum_{\gamma} |t_{l\gamma} t_{m\gamma} \rangle$ respectively. Due to spin rotational invariance we may focus on the total-$S^z = 0$ components only, i.e. $|lm, P\rangle = |lm, z\rangle$ (triplet) and $|lm, D\rangle = |lm, zz\rangle$ (quintet). The bare two-particle eigenstates are

$$|kq, P\rangle = \frac{1}{N} \sum_{l,m} e^{i[k(l+m)/2+q(l-m)]} |lm, P\rangle$$

$$|kq, D\rangle = \frac{1}{\sqrt{N(N-1)}} \sum_{l,m} e^{i[k(l+m)/2} \text{sgn}(l-m) \sin(q(l-m)) |lm, D\rangle$$

where identical properties regarding the momenta as in (3) apply and the bare two-particle eigenstates of the states $|kq, P\rangle$ and $|kq, D\rangle$ are identical to $\langle kq |$ of (3). Note that both, $P$- and $D$-states conform with the hardcore constraint $|t_{l\alpha} t_{l\beta} \rangle = 0$. From the identity $(S_l + S_m)^2 / 2 - 2 = S_l \cdot S_m$ and from (12) it follows directly that

$$\langle kq, X | H_2 | k'q', X \rangle = -\frac{c_X}{N-1} \delta_{kk'} \sin(q) \sin(q')$$

with $c_X = 4, 2$, and $-2$ for $X = S, P$, and $D$ respectively. Therefore the energies $E_k^n$ of the (anti)bound states in all spin channels are obtained from the zeros of the single equation

$$1 + c_X \lambda G(k, E_k^n) = 0$$

with $G(k, z)$ as in (15) and $c_X$ set according to the total spin. For $X = P(D)$ a(n) (anti)bound state exists for $k \geq 2\pi/3$ for all values of $\lambda$ with $E_k^P(D) = 2 - (+)\lambda/2$. In fig. 1 the dispersion of these (anti)bound states relative to the two-triplet continuum is depicted.

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