[NPHYS-2007-06-00643]

SUPPLEMENTARY MATERIAL for "Spinons and triplons in spatially anisotropic frustrated antiferromagnets" by Masanori Kohno, Oleg A. Starykh, and Leon Balents

Basis

The two-spinon states of a single chain are characterized by two continuous quantum numbers, which can be thought of either as the momenta k_{x1}, k_{x2} of the individual (unbound) spinons, or equivalently, the total momentum $k_x = k_{x1} + k_{x2}$ and (excitation) energy $\epsilon = \epsilon_s(k_{x1}) + \epsilon_s(k_{x2})$ of the pair. The spinon excitation energy is given by the des Cloizeaux-Pearson dispersion relation $\epsilon_s(k) = (\pi J/2)\sin[k]$ [36], which is seen to describe the lower boundary of the two-spinon continuum, $\epsilon_s(k) = \omega_{2,l}(k)$. The excitation energy of the spinon pair is then expressed via the total, k_x, and relative, $q_x = (k_{x1} - k_{x2})/2$, momenta of the pair $\epsilon(k_x, q_x) = \pi J \sin[k_x/2]\cos[q_x]$. Observe that the upper (lower) boundaries of the two-spinon continuum correspond to $q_x = 0$ ($\pm k_x/2$). We find it convenient to describe the two-spinon state of a chain in terms of the total momentum k_x and excitation energy ϵ of the pair. The transformation from q_x to ϵ explains the density of states factor $D_{k_x}(\epsilon)$ appearing in (2).

To derive (3), we evaluate the expectation value of the Hamiltonian (1) in the state (2)

$$\langle\Psi|\mathcal{H}|\Psi\rangle_{\mathbf{k}} = E_0 L_x L_y + \int d\epsilon D_{k_x}(\epsilon)\epsilon|\psi_{k_x}(\epsilon)|^2 + J'(\mathbf{k})\int d\epsilon d\tilde{\epsilon} D_{k_x}(\epsilon)D_{k_x}(\tilde{\epsilon})A^*_{k_x}(\epsilon)A_{k_x}(\tilde{\epsilon})\psi^*_{k_x}(\epsilon)\psi_{k_x}(\tilde{\epsilon}), \tag{7}$$

where $E_0 = J(-\ln 2 + 1/4)$ is the ground state energy per site of decoupled chains [34, 35], and we made use of the normalization condition $\int d\epsilon D_{k_x}(\epsilon)|\psi_{k_x}(\epsilon)|^2 = 1$ as appropriate for the state (2). "Factoring out" ϵ-integration $\int d\epsilon D_{k_x}(\epsilon)\psi^*_{k_x}(\epsilon)$ in (7) leads to Eq.(3) of the main text.

The ground state to two spinon matrix element $A_{k_x}(\epsilon)$ represents the key technical element of our calculation

$$A_{k_x}(\epsilon) \equiv \sqrt{2}\langle 0|S^x_{-k_x,y}|k_x,\epsilon\rangle_y = -i\sqrt{2}\langle 0|S^y_{-k_x,y}|k_x,\epsilon\rangle_y. \tag{8}$$

Its absolute value squared, $M(k_x,\epsilon) = |A_{k_x}(\epsilon)|^2$, also called the singlet-to-triplet transition rate, is obtained exactly by an algebraic analysis based on infinite-dimensional quantum group symmetries in Ref. [38] and subsequently simplified in Ref. [39], which we follow here. The matrix element A in (8) is obtained as a square-root of M because two-spinon states with different k_x and/or ϵ are orthogonal and thus the phase can be set to zero for every set (k_x,ϵ) independently. Note that our choice of $|k_x,\epsilon_{k_x}\rangle_y$ as an $S^z = +1$ eigenstate of 2 spinons in y-th chain ensures that $\langle 0|S^z_{-k_x,y}|k_x,\epsilon\rangle_y = 0$.

Specifically, $A_k(\epsilon) = \exp[-I(t)/2]/\sqrt{4\pi}$, where [39]

$$I(t) = -I_0 - \ln|t\sinh(\pi t/4)| + s(t)\ ,\ s(t) = \int_0^\infty dx \frac{\sin^2[xt/2]}{x\cosh^2[x]}, \tag{9}$$

$I_0 = 0.3677...$ and k,ϵ dependence comes via the parameter t

$$\cosh[\frac{\pi t}{4}] = \sqrt{\frac{\omega^2_{2,u}(k) - \omega^2_{2,l}(k)}{\epsilon^2 - \omega^2_{2,l}(k)}}. \tag{10}$$

Integration of $s(t)$ has been performed numerically by an adaptive quadrature algorithm.

Discretization in the ϵ space

For numerical calculations, we carefully discretize energy ϵ for every k_x value, by dividing the interval $\omega_{2,u}(k_x) - \omega_{2,l}(k_x)$ into M discrete points. The resulting discrete $M \times M$ eigenvalue problem is then solved for every value of momentum $\mathbf{k} = (k_x, k_y)$. The data points in the ϵ-space are chosen so that the distribution of them reduces to the exact density of states of the Heisenberg chain [37] in the continuous limit:

$$\Delta\epsilon D_{k_x}(\epsilon) = \frac{|k_x|}{2M}. \tag{11}$$

Next, it is convenient to define rescaled two-spinon states with Kronecker delta function overlaps in place of the Dirac delta-function overlapping continuum states. Specifically:

$$|\epsilon\rangle^M \equiv \sqrt{\frac{|k_x|}{2M}}|\epsilon\rangle. \tag{12}$$

One can check that

$$ {}^{M}\langle\epsilon'|\epsilon\rangle^{M} = \frac{k_x}{2M}\langle\epsilon'|\epsilon\rangle = \frac{k_x}{2M}\frac{1}{D(\epsilon)}\delta(\epsilon-\epsilon') = \frac{k_x}{2M}\frac{1}{D(\epsilon)\Delta\epsilon}\delta_{\epsilon,\epsilon'} = \delta_{\epsilon,\epsilon'} \tag{13} $$

as desired. We also rescale wavefunction as $\phi(\epsilon) = \sqrt{|k_x|/(2M)}\psi_{\mathbf{k}}(\epsilon)$, suppressing for brevity its dependence on the center-of-mass momentum $\mathbf{k}$ (it enters the problem only as a parameter). With these definitions, equation (2) takes on simple form

$$ |\Psi_{\mathbf{k}}\rangle = \sum_{\epsilon}\phi(\epsilon)|\epsilon\rangle^{M}. \tag{14} $$

The Schrödinger equation, in turn, takes on matrix form

$$ \epsilon\phi(\epsilon) + J'(\mathbf{k})\frac{|k_x|}{2M}\sum_{\tilde{\epsilon}}A^{*}(\tilde{\epsilon})A(\epsilon)\phi(\tilde{\epsilon}) = E_{\mathbf{k}}\phi(\epsilon). \tag{15} $$

We typically took the number of discretized energies M to be several thousand, as large as necessary to ensure good resolution and convergence of the results.

Dynamical structure factor $S(k,\omega)$

The dynamical structure factor $S(\mathbf{k},\omega)$ is defined by

$$ S(\mathbf{k},\omega) \equiv \int\frac{dt}{2\pi}\mathrm{e}^{\mathrm{i}\omega t}\langle\mathrm{G.S.}|S^{\alpha}_{-\mathbf{k}}(t)S^{\alpha}_{\mathbf{k}}(0)|\mathrm{G.S.}\rangle, \tag{16} $$

where *no sum* on $\alpha = x, y$ or z is implied. This is calculated within the 2-spinon subspace using the obtained eigenstates and energies of the effective Hamiltonian as

$$ S(\mathbf{k},\omega) = \frac{1}{2}\sum_{E_{\mathbf{k}}}|\langle\mathrm{G.S.}|S^{-}_{-\mathbf{k}}|\Psi_{\mathbf{k}}\rangle|^{2}\delta(\omega-E_{\mathbf{k}}), \tag{17} $$

where summation is over excited states of the system with momentum $\mathbf{k}$ and energy $\omega = E_{\mathbf{k}}$. In Eq.(17), it is clear that, since the ground state is a singlet, only triplet excited states $|\Psi_{\mathbf{k}}\rangle$ with total $S^{z} = +1$ can contribute to the sum.

Note that

$$ |\Psi_{\mathbf{k}}\rangle = \sum_{\epsilon}\frac{1}{\sqrt{L_y}}\sum_{y}\phi(\epsilon)e^{ik_y y}|k_x,\epsilon,y\rangle^{M}, \tag{18} $$

and writing $S^{-}_{-\mathbf{k}} = \frac{1}{\sqrt{L_y}}\sum_{y'}e^{-ik_y y'}S^{-}_{-k_x,y'}$, we have

$$ S^{-}_{-\mathbf{k}}|\Psi_{\mathbf{k}}\rangle = \sum_{\epsilon}\frac{1}{L_y}\sum_{y,y'}\phi(\epsilon)e^{ik_y(y-y')}S^{-}_{-k_x,y'}|k_x,\epsilon,y\rangle^{M}. \tag{19} $$

This can be separated into terms with $y' = y$ and $y' \neq y$. Projecting the resulting state into the subspace containing only zero or two spinons per chain, one then obtains

$$ S^{-}_{-\mathbf{k}}|\Psi_{\mathbf{k}}\rangle = |\Upsilon\rangle_0 + |\Upsilon\rangle_1, \tag{20} $$

with components containing zero or four total spinon excitations:

$$ |\Upsilon\rangle_0 = \sqrt{2}\sqrt{\frac{|k_x|}{2M}}\sum_{\epsilon}\phi(\epsilon)A_{k_x}(\epsilon)|\mathrm{G.S.}\rangle_0, \tag{21} $$

$$ |\Upsilon\rangle_1 = \sum_{\epsilon,\epsilon'}\sqrt{2}\sqrt{\frac{|k_x|}{2M}}\frac{1}{L_y}\sum_{y\neq y'}e^{ik_y(y-y')}\phi(\epsilon)A^{*}_{-k_x}(\epsilon')|k_x,\epsilon;S^{z}=+1\rangle^{M}_{y}|-k_x,\epsilon';S^{z}=-1\rangle^{M}_{y'}\otimes_{y''\neq y,y'}|0\rangle_{y''}. \tag{22} $$

Note that in Eq.(22) we have indicated explicitly the total S^z of the spinon pair on chains y, y', since the two chains have equal and opposite $S^z = \pm 1$.

We approximate the ground state by its form to first order in $J'(\mathbf{k})$, in the subspace of states with zero or two spinons per chain:

$$|\text{G.S.}\rangle \approx |\text{G.S.}\rangle_0 + |\text{G.S.}\rangle_1, \tag{23}$$

with as usual, in first order perturbation theory,

$$|\text{G.S.}\rangle_1 = \frac{1}{E_0 - \mathcal{H}_0}\mathcal{H}'|\text{G.S.}\rangle_0. \tag{24}$$

Here $\mathcal{H}_0 = \mathcal{H}|_{J'_a \to 0}$ is the decoupled chain Hamiltonian, and $\mathcal{H}' = \mathcal{H} - \mathcal{H}_0$ contains the interchain exchange couplings.

The desired matrix element then has two terms:

$$\langle \text{G.S.}|S^-_{-\mathbf{k}}|\Psi_{\mathbf{k}}\rangle = {}_0\langle \text{G.S.}|\Upsilon\rangle_0 + {}_1\langle \text{G.S.}|\Upsilon\rangle_1. \tag{25}$$

The first term is simplest. From Eq.(21), one directly obtains

$${}_0\langle \text{G.S.}|\Upsilon\rangle_0 = \sqrt{2}\sqrt{\frac{|k_x|}{2M}}\sum_\epsilon \phi(\epsilon)A_{k_x}(\epsilon). \tag{26}$$

Now consider the second term. To evaluate this explicitly, it is useful to write

$$\mathcal{H}' = \sum_{k_x,y} J'(k_x)\left[\frac{1}{2}\left(S^+_{k_x,y}S^-_{-k_x,y+1} + S^-_{k_x,y}S^+_{-k_x,y+1}\right) + S^z_{k_x,y}S^z_{-k_x,y+1}\right]. \tag{27}$$

Because $|\Upsilon\rangle_1$ contains only spin $S^z = \pm 1$ spinon pairs on two chains, we can restrict the consideration of $|\text{G.S.}\rangle_1$ to those components with the same structure. This means that the third term inside the square brackets in Eq.(27) can be neglected, since it creates $S^z = 0$ two-spinon states on the chains $y, y+1$. Taking only the first two terms, we have

$$|\text{G.S.}\rangle_1 = -\sum_{k_x,y}\frac{|k_x|}{2M}J'(k_x)\sum_{\epsilon,\epsilon'}\frac{A^*_{k_x}(\epsilon)A^*_{-k_x}(\epsilon')}{\epsilon+\epsilon'}\sum_{\delta=\pm 1}|k_x,\epsilon;S^z=\delta\rangle^M_y|-k_x,\epsilon';S^z=-\delta\rangle^M_{y+1}\otimes_{y''\neq y,y+1}|0\rangle_{y''}. \tag{28}$$

We can then evaluate the overlap. One obtains

$${}_1\langle \text{G.S.}|\Upsilon\rangle_1 \;=\; -\sqrt{2}\left(\frac{|k_x|}{2M}\right)^{3/2}J'(\mathbf{k})\sum_{\epsilon,\epsilon'}\frac{\phi(\epsilon)A_{k_x}(\epsilon)A^*_{-k_x}(\epsilon')A_{-k_x}(\epsilon')}{\epsilon+\epsilon'}. \tag{29}$$

Observe that the correction diverges at $k_x = \pi$, where the spinons have zero energy, *unless* $J'(k_x = \pi) = 0$. This hints at the instability of the system towards magnetic ordering in the case of *non-frustrated* inter-chain coupling.

Combining Eqs.(29,26), the perturbation-theory- improved transition rate can be expressed as

$$|\langle \text{G.S.}|S^-_{-\mathbf{k}}|\Psi_{\mathbf{k}}\rangle|^2 = \frac{|k_x|}{M}\left|\sum_\epsilon \phi(\epsilon)A_{k_x}(\epsilon)\left\{1 - J'(\mathbf{k})\sum_{\epsilon'}\frac{|k_x|}{2M}\frac{|A_{k_x}(\epsilon')|^2}{\epsilon+\epsilon'}\right\}\right|^2. \tag{30}$$

This and equation (17) provides way to numerically evaluate the structure factor. Fig. 5 shows that effect of the perturbative correction ${}_1\langle \text{G.S.}|\Upsilon\rangle_1$ on the calculated structure factor is very small.

Connection to random-phase approximation (RPA)

It is possible to obtain an analytic solution to the integral equation describing the two-spinon states, Eq.(3). To do so, it is convenient to use the same discretization scheme as described above in Eq.(11) to resolve the wavefunctions of individual states in the continuum. The Schrödinger equation (15) is re-written as

$$\epsilon\phi(\epsilon) + B(E)A(\epsilon) = E\phi(\epsilon). \tag{31}$$

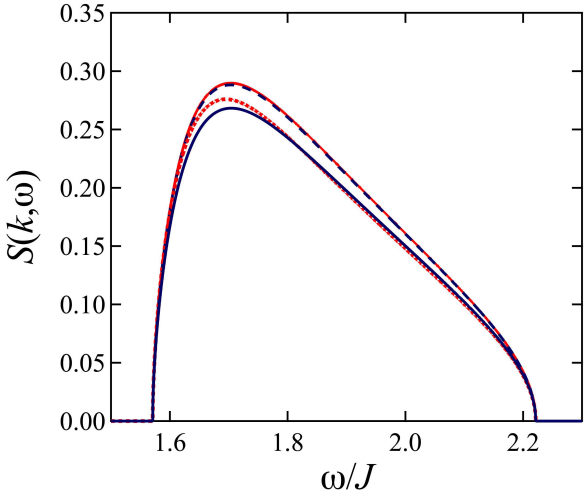


FIG. 5: Comparison of $S(\mathbf{k},\omega)$ at $\mathbf{k}' = (-\pi/2, 4\pi)$ between the present approximation (17) and (30) (blue solid line), that without the correction to the ground state (blue dashed line), RPA as derived in (4), (48) and (49) (red solid line), and the standard RPA (50) (red dotted line).

Note that the quantity

$$B(E) = \frac{J'(\mathbf{k})|k_x|}{2M} \sum_{\tilde{\epsilon}} \phi(\tilde{\epsilon}) A^*(\tilde{\epsilon}) \tag{32}$$

is *independent* of ϵ. This allows one to completely determine the ϵ dependence of $\phi(\epsilon)$ as

$$\phi_E(\epsilon) = \frac{B(E)A(\epsilon)}{E-\epsilon}. \tag{33}$$

Inserting this form into Eq.(32), one obtains the eigenvalue condition

$$\frac{J'(\mathbf{k})|k_x|}{2M} \sum_{\epsilon} \frac{|A(\epsilon)|^2}{E-\epsilon} = 1. \tag{34}$$

This equation has two classes of solutions. There are bound (or anti-bound) states, in which the eigenvalue E is well-separated from the continuum of the decoupled chains. Then the denominator in Eq.(34) remains finite as one takes the discretization to zero, i.e. $M \to \infty$. On thereby obtains the bound state condition

$$J'(\mathbf{k}) \int d\epsilon\, D(\epsilon) \frac{|A(\epsilon)|^2}{E-\epsilon} = 1. \tag{35}$$

The second class of solution is more subtle, and occurs when E is in the range of the continuum, i.e. for finite but large M, it is close to one of the discretized two-spinon eigenvalues of the decoupled chains, which we denote ϵ_0. We assume (and confirm self-consistently) that the energy of such a state can be written as $E = \epsilon_0 + \delta/M$, where δ remains $O(1)$ as $M \to \infty$. We then rewrite Eq.(34) as

$$\frac{J'(\mathbf{k})|k_x|}{2M} \sum_{\epsilon} |A(\epsilon)|^2 \left[\frac{\epsilon_0 - \epsilon}{(\epsilon_0-\epsilon)^2 - (\delta/M)^2} - \frac{\delta/M}{(\epsilon_0-\epsilon)^2 - (\delta/M)^2} \right] = 1. \tag{36}$$

The first term contains $\epsilon_0 - \epsilon$ in the *numerator*, and the summand is locally *odd* about $\epsilon = \epsilon_0$. The contribution from the sum in the region where $|\epsilon - \epsilon_0|$ is $O(1/M)$ is therefore negligible, because the contributions from eigenvalues ϵ on opposite sides of ϵ_0 cancel. Conversely, there is a non-vanishing contribution from $|\epsilon - \epsilon_0|$ of $O(1)$, which in the $M \to \infty$ limit becomes a principle part integral. In the second sum, there is only a δ/M factor in the numerator, and the integrand is locally *even* about $\epsilon = \epsilon_0$. In this sum, there is an $O(1)$ contribution from the region $|\epsilon - \epsilon_0|$ of $O(1/M)$. This must be calculated explicitly, by summing over discrete $\epsilon_n = \epsilon_0 + \Delta\epsilon n$, with integer n. Here the level spacing is determined from $D(\epsilon_0)\Delta\epsilon = |k_x|/(2M)$, and because the sum is sharply peaked we can approximate

$A(\epsilon) \approx A(\epsilon_0)$. Because the integrand decays as $|\epsilon - \epsilon_0|^{-2}$, the contribution from $|\epsilon - \epsilon_0|$ of $O(1)$ is negligible in the sum, and the limits on n can be extended to $\pm\infty$.

Carrying out this sum (using $2b^2 \sum_1^\infty 1/(n^2 - b^2) = 1 - \pi b \cot(\pi b)$) and taking the $M \to \infty$ limit, we find the simple result

$$\pi J'(\mathbf{k}) D(\epsilon_0) |A(\epsilon_0)|^2 \cot(\pi\gamma) + J'(\mathbf{k}) P \int d\epsilon \, \frac{D(\epsilon)|A(\epsilon)|^2}{\epsilon_0 - \epsilon} = 1, \tag{37}$$

where we have defined for convenience

$$\gamma = \frac{2D(\epsilon_0)\delta}{|k_x|} = \frac{\delta E}{\Delta\epsilon}. \tag{38}$$

This form guarantees $|\gamma| < 1/2$, so that the shift of the energy level δE is always less than half the distance to the nearest eigenvalue, i.e. the levels do not cross upon increasing J'.

Now we turn to the determination of the structure factor. Normalization of state (14) fixes $|B(E)|$ according to

$$|B(E)|^2 = \left[\sum_\epsilon \frac{|A(\epsilon)|^2}{(E-\epsilon)^2} \right]^{-1}. \tag{39}$$

This observation leads us to the structure factor, which we divide into the bound state and continuum contributions:

$$S(\mathbf{k}, \omega) = S_{\rm bs}(\mathbf{k}, \omega) + S_{\rm cont}(\mathbf{k}, \omega). \tag{40}$$

First consider the bound (anti-bound) state contribution – we will consider only a single bound (anti-bound) state, since this occurs for the triangular lattice of present interest. In any case, multiple states would simply give additive contributions. This gives directly a delta-function peak in the structure factor, at $\omega = E_{bs}$, where E_{bs} is the bound state energy:

$$S_{\rm bs}(\mathbf{k}, \omega) = |B(E)|^2 \frac{|k_x|}{2M} \sum_{\epsilon, \epsilon'} \frac{|A(\epsilon)|^2 |A(\epsilon')|^2}{(E-\epsilon)(E-\epsilon')} \delta(\omega - E). \tag{41}$$

Using Eq.(34), this immediately simplifies to

$$S_{\rm bs}(\mathbf{k}, \omega) = |B(E)|^2 \frac{2M}{|k_x|} \frac{1}{[J'(\mathbf{k})]^2} \delta(\omega - E). \tag{42}$$

In this case, since $E - \epsilon$ remains non-zero as $M \to \infty$, the sum in Eq.(39) can be converted to an integral:

$$|B(E)|^2 = \frac{|k_x|}{2M} \left[\int d\epsilon \, D(\epsilon) \frac{|A(\epsilon)|^2}{(E-\epsilon)^2} \right]^{-1}. \tag{43}$$

Thus we obtain the bound state delta-function contribution

$$S_{\rm bs}(\mathbf{k}, \omega) = \left\{ [J'(\mathbf{k})]^2 \int d\epsilon \, D(\epsilon) \frac{|A(\epsilon)|^2}{(E_{bs} - \epsilon)^2} \right\}^{-1} \delta(\omega - E_{bs}). \tag{44}$$

We now turn to the continuum contribution. Following the same steps as above, we find the analog of Eq.(42),

$$S_{\rm cont}(\mathbf{k}, \omega) = \sum_{E \neq E_{bs}} |B(E)|^2 \frac{2M}{|k_x|} \frac{1}{[J'(\mathbf{k})]^2} \delta(\omega - E). \tag{45}$$

In this case, more care must be taken in evaluating $B(E)$ from Eq.(39), because the energy denominators in the sum become small as $M \to \infty$. Indeed, the sum is dominated by $|E - \epsilon|$ of $O(1/M)$, and so one may as in Eq.(37) consider the density of states (i.e. spacing $\Delta\epsilon$) and $A(\epsilon)$ to be approximately constant in this region. This allows one to carry out the sum explicitly (using $\sum_{-\infty}^{\infty} 1/(n+a)^2 = \pi^2/\sin^2(\pi a)$) and obtain

$$|B(E)|^2 = \frac{(\Delta\epsilon)^2}{\pi^2 \csc^2(\pi\gamma)} \frac{1}{|A(E)|^2}. \tag{46}$$

Inserting this into Eq.(45), in the large M limit the sum may be converted to an integral via $\sum_{E\neq E_{bs}} \Delta E \to \int dE$, and thereby collapse the delta-function. One then obtains

$$S_{\rm cont}(\mathbf{k},\omega) = \frac{1}{\pi^2 \csc^2(\pi\gamma)} \frac{1}{D(\omega)|A(\omega)|^2} \frac{1}{[J'(\mathbf{k})]^2}. \tag{47}$$

Using $\csc^2(\pi\gamma) = \cot^2(\pi\gamma) + 1$ and Eq.(37), we finally arrive at

$$S_{\rm cont}(\mathbf{k},\omega) = \frac{S_{1D}(k_x,\omega)}{[1 + J'(\mathbf{k})\chi'(k_x,\omega)]^2 + [J'(\mathbf{k})\chi''(k_x,\omega)]^2} \tag{48}$$

with

$$\chi'(k_x,\omega) = P\int_0^\infty d\omega' \, \frac{D(\omega')|A(\omega')|^2}{\omega' - \omega} \text{ and } \frac{1}{\pi}\chi''(k_x,\omega) = S_{1D}(k_x,\omega) = D(\omega)|A(\omega)|^2. \tag{49}$$

The standard RPA result has the same functional form as (48) but with χ', χ'' replaced by $\mathrm{Re}\chi_{\rm rpa}, \mathrm{Im}\chi_{\rm rpa}$ via

$$\mathrm{Re}\chi_{\rm rpa}(k_x,\omega) = \frac{1}{\pi}P\int_{-\infty}^\infty d\omega' \, \frac{\mathrm{Im}\chi_{\rm rpa}(k_x,\omega')}{\omega' - \omega} \text{ and } \frac{1}{\pi}\mathrm{Im}\chi_{\rm rpa}(k_x,\omega) = \mathrm{sgn}(\omega) S_{1D}(k_x,|\omega|). \tag{50}$$

The close similarity of expressions (49) and (50) is illustrated in Fig.5.

Triplon dispersion in anisotropic triangular lattice

Bound (and antibound) states outside the continuum are determined by the condition $1 + J'(\mathbf{k})\chi'(k_x,\omega_t(\mathbf{k})) = 0$, which is just (35), where $J'(\mathbf{k}) = 4J'\cos[k_x/2]\cos[k_y/2]$ for Cs_2CuCl_4.

Bound state: Since $\chi' > 0$ for $\omega < \omega_{2,l}$ as follows from (49), one needs $J'(k_x,k_y) < 0$ for it to appear. Using the asymptotic behavior of the chain structure factor near the lower edge of the two-spinon continuum, $\omega_{2,l}$ [39] $S_{1D} \approx C\sqrt{-\ln[\omega - \omega_{2,l}]/[\omega_{2,l}(\omega - \omega_{2,l})]}$, we find with logarithmic accuracy

$$\chi'(k_x,\omega\to\omega_{2,l}) = \sqrt{8}C \frac{\sqrt{-\ln[\omega_{2,l} - \omega]}}{\sqrt{\omega_{2,l}[\omega_{2,l} - \omega]}} \arctan\sqrt{\frac{\omega_{2,u} - \omega_{2,l}}{\omega_{2,l} - \omega}}. \tag{51}$$

Here $C = \exp[I_0/2]/\sqrt{16\pi^3}$. Provided that $J'(\mathbf{k})$ is negative, we readily see that the triplon binding energy behaves as $\delta E = \omega_{2,l} - \omega_t(\mathbf{k}) \propto [J'(\mathbf{k})]^2$, up to very weak logarithmic corrections. The triplon appears below the continuum and propagates along both the x and y directions, as shown in Fig.2a.

To compare with the bound state data from the effective Schrödinger equation (3), we calculate S_{1D} and χ' numerically. Results for the binding energy $\delta E = \omega_{2,l} - \omega_t$ obtained in these two calculations are found to essentially coincide with each other as shown in Fig. 6. We observe that in the regions where the width of the 2-spinon continuum becomes comparable to δE, the scaling changes to $\delta E \sim J'(\mathbf{k})$. The change from quadratic to linear scaling can be understood from (51), and simply corresponds to the situations where the argument of the cotangent is large (quadratic) or $O(1)$ (linear). We also analyze spectral weight (residue) Z of the triplon, see (44). We find that for small J' it scales as $\sqrt{\delta E} \propto J'$. A comparison between the numerical and analytical results in a wider range of inter-chain exchange values is shown in Fig.6.

It should be mentioned that the multiplicative logarithmic factor in χ', eq.(51), does lead to a weak "spiral" instability at zero frequency – i.e. in this approach one finds a bound state with negative energy at some ordering momentum (measured from π)[41]. This instability, however, is *very* weak. The corresponding ordering momentum is extremely small, $k_{x,0} \sim 10^{-10}$ [41], translating into similarly tiny "instability energy" $\sim Jk_{x,0}$. Moreover, this classical instability is overshadowed by a stronger quantum (of collinear type) ones, of the order $(J')^4/J^3$ [31]. Since our approximation is concerned with the features of the dynamical structure factor at energies of order J' and higher, we are allowed to disregard these weak instabilities.

Anti-bound states are analyzed similarly. In this case, $\omega_t(\mathbf{k}) = \omega_{2,u}(k_x) + \delta_t(\mathbf{k})$ is above the 2-spinon continuum where $\chi' < 0$, see (49). Since $\chi'(\omega)$ continuously decreases in magnitude to zero but retains the same (negative) sign as ω in increased from ω_{2u} to ∞, the condition for the existence of an anti-bound state is simply $1 + J'(\mathbf{k})\chi'(k_x,\omega_{2,u}) < 0$. The anti-bound state therefore merges into the two-spinon continuum when $1 + J'_{\rm crit}(\mathbf{k})\chi'(k_x,\omega_{2,u}) = 0$. As $J'(\mathbf{k})$ is increased above this threshold, one can show that, because $S(\mathbf{k},\omega) \sim \mathrm{Const}\sqrt{\omega_{2u} - \omega}$ for $\omega \lesssim \omega_{2u}$ [39], the anti-bound state energy scales as $\delta_t(\mathbf{k}) \propto (J'(\mathbf{k}) - J'_{\rm crit}(\mathbf{k}))^2$ while its spectral weight scales as $\sqrt{\delta_t}$, similarly to

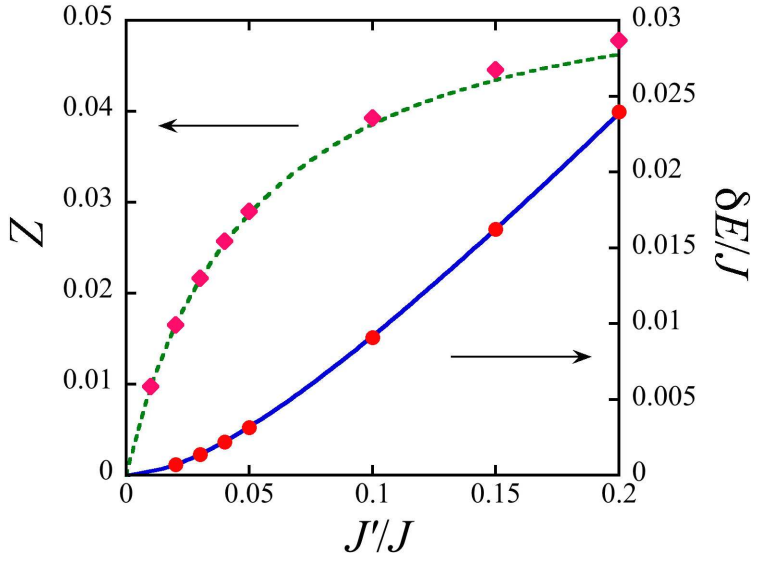


FIG. 6: (Left y-axis) Transition rate to the bound state (Z). Pink diamonds are obtained by the present approach, and the green dotted line is the analytical result in Eq.(44). The small deviation in the large J'/J regime is due to the correction to the ground state which is not included in Eq.(44). (Right y-axis) Gap between the bound state and the lower edge of continuum (δE). Red circles are obtained by the present approach, and the blue solid line denotes the RPA result. The data shown here are calculated at $\mathbf{k} = (\pi/4, \pi)$ on a spatially anisotropic triangular lattice with $J_1' = J_2' (\equiv J')$ and $J_3' = 0$.

the bound state situation discussed above. Unlike the bound state, the anti-bound one is not a completely sharp excitation. This is because it takes place in the region of 4-spinon excitations, which extend from $\omega_{2,l}(k_x)$ up to $\omega_{4,u}(k_x) = \pi J\sqrt{2(1+|\cos[k_x/2]|)}$, [40]. Hence, $\chi'' \neq 0$ and the triplon lineshape is in fact Lorentian, see (48). However, the 4-spinon spectral weight is very small in the region between $\omega_{2,u}$ and $\omega_{4,u}$ boundaries [40], and we find that anti-bound states remain well defined, with the height-to-width ratio well above 1.

Being a collective excitation above the two-particle continuum, the anti-bound triplon here is very similar to the familiar zero-sound mode in a Fermi-liquid, e.g. ^{3}He. The analogy is made much more precise by focusing on the region near Γ point in the Brillouin zone, where the 2-spinon continuum collapses onto a line: $\omega_{2,u} - \omega_{2,l} \sim k_x^3$ as $k_x \to 0$. In this region $\chi'(k_x, \omega) = |k_x|/(2(v_s|k_x| - \omega))$, where $v_s = \pi J/2$ is spinon velocity. The dispersion is found immediately (see Fig.2b):

$$\omega_t(\mathbf{k}) = v_s|k_x|\Big(1 + \frac{J'(\mathbf{k})}{2v_s}\Big) \text{ for } J'(\mathbf{k}) > 0. \tag{52}$$

We see that the anti-bound triplon is just an "acoustic plasmon" of the spinon gas with short-range interactions.

As anti-bound states away from the Γ point require strong J' for their existence, we would like to suggest that somewhat more two-dimensional spatially anisotropic triangular antiferromagnet Cs_2CuBr_4 [50] seems to be a promising candidate for the corresponding inelastic neutron scattering study.