Adjacent spin operator correlations in the Heisenberg spin chain
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Chapter 6

Spin-exchange correlation function

In this chapter we use the previous results for the $S^{4z}(q,\omega)$ in order to calculate the spin-exchange dynamical structure factor which is directly related to RIXS measurement in the isotropic spin chain. We then compare a numerical evaluation of this correlation function with the single spin DSF $S^{zz}(q,\omega)$, which is proportional to the INS cross section.

6.1 Dynamical structure factor and total spin sectors

With the spin exchange operator defined in (4.14)

$$X_q = \frac{1}{\sqrt{N}} \sum_j e^{iqj} (S_{j-1} \cdot S_j + S_j \cdot S_{j+1}) ,$$

(6.1)

the spin-exchange DSF is defined as

$$S^{\text{exch.}}(q,\omega) = \frac{2\pi}{N} \sum_{\alpha} \left| \langle \text{GS} | \sum_j e^{iqj} (S_{j-1} \cdot S_j + S_j \cdot S_{j+1}) |\alpha\rangle \right|^2 \delta(\omega - \omega_{\alpha})$$

$$= \frac{4\pi(1 + \cos(q))}{N} \sum_{\alpha} \left| \langle \text{GS} | \sum_j e^{-iqj} S_j \cdot S_{j+1} |\alpha\rangle \right|^2 \delta(\omega - \omega_{\alpha})$$

(6.2)

where the normalized ground state is $|\text{GS}\rangle$, the normalized excited states $|\alpha\rangle$, excitation energies $\omega_{\alpha} = E_{\alpha} - E_{\text{GS}}$. By expanding explicitly the spin operator inner
product, the DSF contains
\[
S_{\text{exch.}}(q, \omega) = \frac{8 \pi \cos^2(q/2)}{N} \sum_{\alpha} \delta(\omega - \omega_{\alpha}) 
\sum_{a, b=x, y, z} \sum_{i, j} e^{-iq(i-j)} \langle GS| S_i^a S_{i+1}^a |\alpha\rangle \langle \alpha| S_i^b S_{i+1}^b |GS\rangle . \tag{6.3}
\]

In the specific case of the isotropic spin chain in zero magnetic field, we can exploit the spin isotropy of the system to express the full spin-exchange dynamical structure factor as a function of the $S_i^z S_{i+1}^z$ form factors only. By globally rotating $\langle 0| S_j^x S_{j+1}^x |\alpha\rangle$ and $\langle 0| S_j^y S_{j+1}^y |\alpha\rangle$ about the $y$ and $x$ axes, respectively, and using the fact that the ground state is a global $su(2)$ singlet, one can show that only singlet excited states contribute to (6.2).

Noticing that the spin-exchange operator conserves $S_{\text{tot}}^z$:
\[
\sum_{i, j} e^{-iq(i-j)} [S_i^z, S_i \cdot S_{i+1}] = 0
\]

one finds that the only contributing $|\alpha\rangle$ are of $S_{\text{tot}}^z = 0$ and by the Wigner-Eckart theorem, similarly to (5.55), $|\alpha\rangle$ can only belong to total spin $S_{\text{tot}} = 0$ or 2. Let’s denote the states of these two sectors by $|\alpha_{S_{\text{tot}}=0,2}\rangle$. The singlet states are invariant under rotation and therefore $e^{i\phi S^a}|\alpha_0\rangle = |\alpha_0\rangle$ with $a = x, y, z$. Then the expansion of the rotation about the $y$ or $x$ axis for the states of $S_{\text{tot}} = 2$ reads
\[
e^{i\phi S^y}|\alpha_2\rangle = \sum_{n=0}^{\infty} \left(\frac{-\phi}{2}\right)^n \frac{1}{n!} (S^+ - S^-)^n |\alpha_2\rangle
\]
\[
e^{i\phi S^x}|\alpha_2\rangle = \sum_{n=0}^{\infty} \left(\frac{i\phi}{2}\right)^n \frac{1}{n!} (S^+ + S^-)^n |\alpha_2\rangle
\]

with $S^{x, y, \pm}_i = \sum_j S^{x, y, \pm}_i$. As the rotated states must have $S_{\text{tot}}^z = 0$, we can restrict the sum to only even exponent $n$ and with
\[
[S^+, S^-] = 2S^z, \quad (S^+)^3|\alpha_2\rangle = (S^-)^3|\alpha_2\rangle = 0
\]

one can write the intermediate result
\[
(S^+ \pm S^-)^n |\alpha_2\rangle = (\pm 1)^{n/2} \frac{2^4 n!}{4^n} |\alpha_2\rangle + \ldots, S_{\text{tot}}^z \neq 0, \quad (n = 2, 4, 6, \ldots) . \tag{6.8}
\]

We can then rewrite the result identical for the two rotations
\[
e^{i\phi S^x \alpha y}|\alpha_2\rangle = |\alpha_2\rangle + \frac{3}{4} \sum_{m=1}^{\infty} (-1)^m (-2\phi)^{2m} \frac{1}{2m!} |\alpha_2\rangle + \ldots, S_{\text{tot}}^z \neq 0
\]
\[
= \left(\frac{1}{4} + \frac{3}{4} \cos(2\phi)\right) |\alpha_2\rangle + \ldots, S_{\text{tot}}^z \neq 0 . \tag{6.9}
\]
The rotation of the form factor around the $y$-axis is therefore

\[
\langle \text{GS} | S^x_i S^x_{i+1} | \alpha \rangle = \langle \text{GS} | e^{i \frac{\pi}{4} S^y_i} S^z_i S^z_{i+1} e^{-i \frac{\pi}{4} S^y_i} | \alpha \rangle = \begin{cases} 
\langle \text{GS} | S^z_i S^z_{i+1} | \alpha \rangle, & S_{\text{tot}} = 0 \\
-\frac{1}{2} \langle \text{GS} | S^z_i S^z_{i+1} | \alpha \rangle, & S_{\text{tot}} = 2. 
\end{cases}
\]

(6.10)

And we have the same result for the rotation of $\langle \text{GS} | S^y_i S^y_{i+1} | \alpha \rangle$ about the $x$-axis.

The spin-exchange DSF can then be expressed exclusively as a function of the $z$-component form factor:

\[
\frac{N \cdot S_{\text{exch}}(q, \omega)}{8 \pi \cos^2(q/2)} = 9 \sum_{\alpha_0} \delta(\omega - \omega_{\alpha_0}) \left| \sum_j e^{-i q j} \langle \text{GS} | S^z_j S^z_{j+1} | \alpha_0 \rangle \right|^2 \\
+ \sum_{\alpha_2} \delta(\omega - \omega_{\alpha_2}) \left| \sum_j e^{-i q j} \langle \text{GS} | S^z_j S^z_{j+1} | \alpha_2 \rangle \right|^2 \\
+ \frac{4}{\omega - \omega_{\alpha_2}} \left( - \frac{1}{2} \right) \left| \sum_j e^{-i q j} \langle \text{GS} | S^z_j S^z_{j+1} | \alpha_2 \rangle \right|^2 \\
+ \frac{4}{\omega - \omega_{\alpha_2}} \left( - \frac{1}{2} \right)^2 \left| \sum_j e^{-i q j} \langle \text{GS} | S^z_j S^z_{j+1} | \alpha_2 \rangle \right|^2.
\]

(6.11)

In this last formula, all contributions of total spin 2 sector vanish and the weight of the correlation is only due to singlet states:

\[
S_{\text{exch}}(q, \omega) = \cos^2(q/2) \frac{72 \pi}{N} \sum_{\alpha \in S_{\text{tot}} = 0} \left| \sum_j e^{-i q j} \langle \text{GS} | S^z_j S^z_{j+1} | \alpha \rangle \right|^2 \delta(\omega - \omega_{\alpha}).
\]

(6.12)

In consequence, the evaluation of the DSF can be done with the form factor expression in (5.51).

### 6.2 Sum Rules

In this section, we calculate the analytical results for the integrated intensity and the first frequency moment ($f$-sum rule) which are used as independent checks for the numerical evaluation of the dynamical structure factor. We give more details about the use of the sum rules in section 5.1.5.
6.2

6.2.1 Integrated intensity

The sum over all the momenta and frequencies of the dynamical structure factors is

\[
\frac{1}{N} \sum_{q=0}^{N-1} \frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega S^{\text{exch.}}(q, \omega) = \frac{1}{N^2} \sum_{q} \sum_{j,j'=1}^{N} e^{-iq(j-j')} \\
\cdot \langle \text{GS} | (S_{j-1} \cdot S_j + S_j \cdot S_{j+1}) (S_{j'-1} \cdot S_{j'} + S_{j'} \cdot S_{j'+1}) | \text{GS} \rangle \\
= \frac{1}{4} - \ln(2) + \frac{9}{8} \zeta(3)
\]

(6.13)

where we used the results in [92] and the following intermediate results valid in the isotropic zero field case:

\[
\frac{1}{N} \sum_{j} \langle \text{GS} | (S_j \cdot S_{j+1}) (S_j \cdot S_{j+1}) | \text{GS} \rangle = \frac{3}{16} - \frac{6}{4N} \sum_{j} \langle \text{GS} | S_j^z S_{j+1}^z | \text{GS} \rangle 
\]

(6.14)

\[
\frac{1}{N} \sum_{j} \langle \text{GS} | (S_{j-1} \cdot S_j) (S_j \cdot S_{j+1}) | \text{GS} \rangle = \frac{3}{4N} \sum_{j} \langle \text{GS} | S_{j-1}^z S_{j+1}^z | \text{GS} \rangle .
\]

(6.15)

6.2.2 First frequency moment

We express the \( f \)-sum rule as a double commutator:

\[
\frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \omega S^{\text{exch.}}(q, \omega) \\
= 4 \cos^2(q/2) \sum_{\mu} (E_0 - E_{\mu}) \left| \langle \text{GS} | \frac{1}{\sqrt{N}} \sum_{j} e^{-iqj} S_j \cdot S_{j+1} | \mu \rangle \right|^2 \\
= \frac{2 \cos^2(q/2)}{N} \sum_{j,j'} e^{-iq(j-j')} \langle \text{GS} | \left[ \sum_{i} S_i \cdot S_{i+1}, S_j \cdot S_{j+1} \right], S_{j'} \cdot S_{j'+1} | \text{GS} \rangle
\]

(6.16)

Using the intermediate results

\[
[S_{j-1} \cdot S_j, S_j \cdot S_{j+1}] = -i \sum_{\alpha,\beta,\gamma} \epsilon^{\alpha\beta\gamma} S_{j-1}^\alpha S_j^\beta S_{j+1}^\gamma \\
\sum_{j} e^{-iqj} \left[ \sum_{i} S_i \cdot S_{i+1}, S_j \cdot S_{j+1} \right] = i(e^{iq} - 1) \sum_{j} e^{-iqj} \sum_{\alpha,\beta,\gamma} \epsilon^{\alpha\beta\gamma} S_{j-1}^\alpha S_j^\beta S_{j+1}^\gamma \\
[S_j^\alpha S_{j+1}^\beta, S_j^\gamma S_{j+1}^\gamma] = 0, \quad \text{for } \alpha \neq \gamma, \beta \neq \gamma
\]

(6.17)
with \( \varepsilon^{\alpha\beta\gamma} \) the Levi-Civita symbol. The first frequency moment reads then

\[
\frac{1}{2\pi} \int_{-\infty}^{\infty} d\omega \omega S_{\text{exch}}(q, \omega) = 6 \sin^2(q) \left[ \left( 1 - 4 \cos^2(q/2) \right) \left( \frac{1}{8} \zeta(3) - \frac{1}{6} \ln 2 \right) + \frac{3}{8} \zeta(3) - \frac{\ln 2}{2} \right]
\]

\[
= 6 \sin^2(q) \left[ 0.0347326 \left( 1 - 4 \cos^2(q/2) \right) + 0.1041977472 \right]. \quad (6.18)
\]

We used during the calculation isotropy and rotational invariance of the correlators, the result here is therefore valid only for isotropic spin chain in zero magnetic field.

### 6.3 Numerical results of the spin-exchange DSF and comparison between RIXS and INS

We evaluate here numerically the spin-exchange DSF at zero magnetic field. The process of computation being the same for all the DSFs in this thesis, the details for the procedure can be found in section 5.1.6. The resulting map of the spin-exchange DSF as defined in (6.12) is presented in figure 6.1(a).

RIXS and INS are two different setups which can probe the dynamics of 1D XXX spin chains in e.g. the compound Sr$_2$CuO$_3$ (see chapter 4). As both response signal are determined by DSFs that we can numerically evaluate via the non perturbative methods explained in this work, we then naturally compare the two results and analyze the specificities of each response. In addition to the computation for spin-exchange DSF (4.13) in figure 6.1(a), we therefore also compute the single spin DSF (4.4) in figure 6.1(b).

Whereas the INS response which includes one spin operator associated which a \( S = 1 \), is dominated by 2-spinon excitations (see table 5.3), we might expect the RIXS response function that includes two spin operators, to be mainly carried by 4-spinon states. However, we notice from the computation that the RIXS excitations splits almost completely into two spinons. The contributions to the sum rule shows that the 2-spinon states cover all but \( 10^{-44}\% \) of the spin-exchange DSF for a chain of 400 sites. Therefore as both response functions are dominated by two spinons, it is not surprising to see that both shapes are very similar and correspond to the 2-spinon spectrum. Although the excitation continuum measured by RIXS coincides with the one probed by INS, the signal has a noticeably different distribution of weight and this difference is partly caused by \( \cos^2(q/2) \). This static prefactor originates from the perturbation of two adjacent exchange couplings in the x-ray scattering process. Excitations are then produced with a typical length \( 2a \) (with \( a \) the lattice spacing). Equivalently, in momentum space, they mainly carry \( \pm \frac{\pi}{2} \) (mod \( 2\pi \)) momentum. This interpretation is very well illustrated by the two figures: for the INS (figure 6.1(b)) the signal is at its highest at the antiferromagnetic wavevector \( q = \pi \) at \( \omega \) close to zero, although the RIXS amplitude...
vanishes there but is concentrated above the continuum threshold at $q = \pi/2$. The profiles at fixed-momentum in figure 6.3 show further differences between RIXS and INS by removing the $\cos^2(q/2)$ pre-factor. We notice that the signal distribution between these two normalized responses are clearly distinct: the RIXS response has a broader distribution in energy and this difference is important enough to be measurable experimentally.

The asymptotic behavior of correlations along the spin chain are well described in the vicinity of $q = \pi$ and $\omega = 0$ by low energy effective theories such as the Luttinger liquid theory. Indeed, in the long distance or time asymptotic regime, the leading and subleading terms of a correlation functions are directly calculated. We find, for example, from bosonization in [96], a formula for the adjacent spin operator correlation functions. The behavior of a DSF singularity at $\omega \sim 0$ (e.g. for the single spin DSF in figure 6.1(b)) could then be compared, after Fourier transform, to the predicted effective field theory form. Either the exponent of the polynomial decay can be confirmed or undefined parameters can be pinned down. However, such comparison is not possible with the RIXS intensity since the signal vanishes precisely at low energy. The spin-exchange operator mainly probes high-energy spectrum and the non-perturbative method we have used here is therefore the only way of describing RIXS so far. Nevertheless, it is important to mention that the non-linear Luttinger liquid method is able to describe the singularity of the single spin DSF at such high-energies (lower boundary of the spectrum, around $q = \pi/2$) [97, 98] and one might expect an equivalent effective field theory result for the spin-exchange DSF.

We have evaluated in this chapter the spin-exchange dynamical structure factor which is proportional to the $K$-edge RIXS intensity. Using the intermediate expression developed in the chapter 5, we have given a non-perturbative expression for the RIXS cross section (4.3).
Figure 6.1: (a) Spin-exchange DSF $S^{\text{exch}}(q, \omega)$ and (b) single spin DSF $S^{zz}(q, \omega)$ for $N = 400$ sites in $\hbar = 0$. The intensity around $q = \pi$ is markedly different in the two cases and the signal of the spin-exchange DSF around $q = \frac{\pi}{2}, \frac{3\pi}{2}$ is enhanced.
Figure 6.2: Fixed momentum profiles of the biased spin-exchange DSF $S^{\text{exch}}(q, \omega)/(\cos^2(q/2))$ (plain) and $S^{zz}(q, \omega)$ (dashed), each normalized to its own sum rule.