Probing strong correlations with light scattering: Example of the quantum Ising model

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Probing Strong Correlations with Light Scattering: the Example of the Quantum Ising model

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In this paper we calculate the nonlinear susceptibility and the resonant Raman cross section for the paramagnetic phase of the ferromagnetic Quantum Ising model in one dimension. In this region the spectrum of the Ising model has a gap m. The Raman cross section has a strong singularity when the energy of the outgoing photon is at the spectral gap $\omega_f \approx m$ and a square root threshold when the frequency difference between the incident and outgoing photons $\omega_i - \omega_f \approx 2m$. The latter feature reflects the fermionic nature of the Ising model excitations.

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I. INTRODUCTION

When photons enter into a strongly interacting medium one expects nonlinear phenomena such as frequency mixing and inelastic light scattering [1],[2]. These effects are interesting by itself, but they can also serve as experimental tools to extract otherwise unaccessible information about strongly correlated dynamics since all these tools probe multi-point dynamical correlation functions which carry much richer information than a simple linear response.

In this paper we will present calculations of three- and four-point dynamical correlation functions for the ferromagnetic Quantum Ising (FQI) model in one spatial dimension (1D) and relate them to two spectroscopic probes: the nonlinear susceptibility and the inelastic scattering cross section of light (Raman scattering). We have chosen the 1D FQI model for three reasons. Firstly, this is a strongly correlated model whose applicability to real materials has been firmly established (see the discussion at the end of the paper). Secondly, this model admits a resonance regime of where nonlinear effects are strongly enhanced. The effectiveness of light probes is somewhat restricted by the fact that the photons carry a very small momentum, but in most strongly correlated systems the strongest correlations occur at finite wave vectors of order of the size of the Brillouin zone. The 1D FQI model represents an exception from this rule. The third factor is a comparative simplicity of the calculations which will allow us to concentrate on the crucial points avoiding cumbersome technical details. We consider FQI as the first integrable model among many where multi-point correlation functions can be calculated and their current calculations present a skeleton of the general scheme for all such models.

The quantum Ising model is described by the Hamiltonian

$$H = \sum_n \Big(- J\sigma^z_n \sigma^z_{n+1} + h\sigma^x_n \Big), \tag{1}$$

where σ^a are the Pauli matrices. This model is one of the best studied models of strongly correlated physics (see [3],[4],[5] for a review). Its exact solution was obtained as early as 1928 by Jordan-Wigner transformation which expresses the spin operators in terms of fermionic creation and annihilation ones. For the reasons explained above we are ingterested in the case $J > 0$ (FQI model). The best condensed matter realization known to date is found in columbite $CoNb_2O_6$ [6],[7],[8]. We will discuss it in Conclusions.

The Ising model (1) may describe not only spins, but any coupled two level systems. If these are spins then operators σ^z_n directly couple to external magnetic field: $\mu_B B^z_n \sigma^z_n$. Alternatively the states of two level system may correspond to positions of electric charges in a double well potential. Then σ^a would be the dipole moment operators; the first term in (1) is the dipole-dipole interaction and the transverse field describes the quantum tunneling between the wells. The interaction of dipole moments with the electric field is given by $pE^z_n\sigma^z_n$ with p being the dipole moment magnitude.

When the dominant interaction is ferromagnetic $J > 0$, the strongest fluctuations take place at zero wave vectors which guarantees a direct coupling to the electromagnetic field creating optimal resonance conditions. The Ising model (1) has two phases depending on the sign of $m = h - J$. The resonance occurs in the paramagnetic phase $m > 0$ when the ground state average of the order parameter $\langle \sigma^z \rangle = 0$. In that case the electromagnetic field has a nonzero matrix element between the ground state and single magnon state.

Raman light scattering is a powerful experimental technique frequently used in condensed matter physics. The measured quantity is the inelastic scattering cross section of photons $R(\mathbf{q},\Omega)$ which contains information about the excitations of condensed matter systems with which the photons interact. The theory of Raman scattering was formulated in the nineteen twenties [9, 10] when the formulae for $R(\mathbf{q},\Omega)$ were derived (see also [11]). A radical simplification of these formulae was suggested in [12] where the resonant part of the Raman cross section was expressed as a particular limit of the four point correlation function of the current operators. This simplification allows one to apply to the problem various techniques of quantum field theory such as Feynman diagram expansion and also simplifies the application of nonperturbative techniques.

Another technique to be discussed is a nonlinear response directly related to the three-point correlation function. The related themes are two-dimensional spectroscopy and spectroscopy with entangled photons [13] ,[14]. A relation of third-order nonlinear optical properties to magnetic interactions was demonstrated in [15].

The Jordan-Wigner transformation transforms FQI into a model of noninteracting noninteracting massive Majorana fermions. In the scaling limit $m << J$ their dispersion becomes relativistic $\epsilon(k)=\sqrt{v^2k^2+m^2},\ \ v\sim J$. In what follows we will set $v=1$ to restore J in the final expressions.

The fact that the excitations of the Ising model do not interact does not make the model trivial. Indeed, since σ^z operators are very nonlocal in terms of the fermions, the electromagnetic field has matrix elements between states with different number of fermionic excitations. Such situation is typical for strongly interacting systems and experimental probes of multipoint correlators are highly suitable to reveal this nonlocality. In the paramagnetic phase of FQI the inelastic processes involve matrix elements with odd number of the Ising fermions with the leading low energy processes being transitions from single- to two-fermion states. The fermionic nature of the excitations is reflected in the fact that the cross section vanishes at the threshold: $R(\Omega)\sim(\Omega-2m)^{1/2}$ (see Eqs. (8,9) below.

II. THE OBSERVABLES

A. The nonlinear susceptibility

is the third derivative of the action with respect to the dynamical magnetic field:

$$\chi^{(3)}_{zzx}=\langle\hat{T}\sigma^z(t_1)\sigma^z(t_2)\sigma^x(t_3)\rangle_{connected}, \tag{2}$$

where $\sigma=\sum_n\sigma^z_n$ (we assume that electromagnetic radiation carries no momentum). In the Ising model such response exists only when the polarization of photons is such that the magnetic field has both z and x components so that the coupling to the magnetic field is described as

$$V=\mu_B\sum_n(B^z\sigma^z_n+B^x\sigma^x_n), \tag{3}$$

The nonlinear susceptibility describes the effects or frequency mixing. As follows from (2), the only nonzero third order response includes two magnetic fields with frequencies $\omega_z,\omega_x-\omega_z$ along the z- and one field with frequency $-\omega_x$ along the x-direction. Our result where we take into account only two magnon production processes, is

$$\chi^{(3)}_{zzx}(\omega_z,-\omega_z+\omega_x,-\omega_x)=C^2\mu_B^3(mJ^3)^{-1/4}\times \Big\{\frac{(\omega_x-2m)\,h\,(\omega_x/2m\ +i\delta)}{(\omega_z-m)\,(\omega_x-\omega_z-m)}+(\omega_{x,z}\to-\omega_{x,z}) + 16m\pi\frac{(\omega_x-\omega_z)\,\omega_z-m^2}{(\omega_z^2-m^2)\,((\omega_x-\omega_z)^2-m^2)}\Big\} \tag{4}$$

where $C\sim 1$ is a numerical constant and (see appendix C)

$$h\,(x)=\int_{-\infty}^{\infty}\left(1+\frac{1}{\cosh\theta}\right)^2\frac{1}{\cosh\theta-x}d\theta.$$

B. Relation between the Raman cross section and the correlation functions

As it was stated above we assume the following spin-photon interaction $V=\sum_n pE_n\sigma^z_n$. Then according to Eqs. (2.21) from [12] the cross section for the light beam polarized along the z-axis is given by the following expression

$$R(\omega_i,\omega_f)=2\pi{\mu_B}^4[(h\omega_i)(h\omega_f)]\frac{\chi_R(\omega_i,\omega_f)}{1-\exp[-\beta(\omega_i-\omega_f)]}, \tag{5}$$

where $\beta=1/T$, ω_i and ω_f are frequencies of the incident and the scattered light. We will consider the $T=0$ limit. Then the function χ is expressed as (see Eqs. 2.30, 2.31 from [12])

$$\chi_R(\omega_i,\omega_f)=\frac{1}{2\pi i}\lim_{\delta_1>\delta_2\to 0} \Big\{\tilde{\Xi}(-\omega_i-i\delta_1,\omega_f+i\delta_2,-\omega_f+i\delta_2,\omega_i-i\delta_1)- \tilde{\Xi}(-\omega_i-i\delta_2,\omega_f+i\delta_1,-\omega_f+i\delta_1,\omega_i-i\delta_2)\Big\}, \tag{6}$$

where $\tilde{\Xi}$ is the Fourier transform of the four-point time ordered correlation function:

$$\langle\,\hat{T}\sigma(t_1)\sigma(t_2)\sigma(t_3)\sigma(t_4)\,\rangle_{connected} \tag{7}$$

Below we will derive the expression for (7) in the paramagnetic phase of model (1) at $T=0$ in the limit $m=h-J<<J$ and will use the result to calculate the Raman cross section (5). For $\omega_i>\omega_f>0$ we obtain the result by substituting Eq. (20) into (6) (see appendix

B)

$$\chi_R(\omega_i,\omega_f) \sim (m/J)^{1/2}\frac{J^3}{m^4}\Big[\frac{G\left(\frac{1}{2m}(\omega_i+\omega_f)\right)}{(\omega_f-m)^2(\omega_i-m)^2} + \frac{G\left(\frac{1}{2m}(\omega_i-\omega_f)\right)}{(\omega_f+m)^2(\omega_i-m)^2}\Big] \quad (8)$$

where

$$G(x) = \Theta(x-1)\frac{(x-1)^{1/2}(x+1)^{5/2}}{x^3}\,. \quad (9)$$

The calculation takes into account only 2-particle intermediate states and hence is valid in the range of frequencies $|\omega_i - \omega_f| < 4m$ when the processes with emission of more than 2 particles do not contribute to the inelastic cross section. The threshold for the inelastic scattering is at $\omega_i \pm \omega_f = 2m$ corresponding to the emission of two fermionic excitations.

III. GREEN'S FUNCTIONS

Below we will do our calculations in the most general form valid for all integrable models and at the end apply the results to the Ising model. We will concentrate on the most difficult case of the four-point function, the calculations of the three-point one are comparatively straightforward.

Let ϕ be a scalar bosonic field (for the Ising model it is the scaling limit of the σ^z). Its Green's functions are defined as time ordered n-point functions

$$\begin{aligned}\tau(\underline{x}) &= \langle\, 0\,|\, T\varphi(x_1)\,\ldots\,\varphi(x_n)|\, 0\,\rangle \\ &= \sum_{perm(x)} \Theta_{1\ldots n}(\underline{t})w(\underline{x})\,,\end{aligned}$$

here $w(\underline{x}) = \langle\, 0\,|\,\varphi(x_1)\,\ldots\,\varphi(x_n)|\, 0\,\rangle$ is the Wightman function and $\Theta_{1\ldots n}(\underline{t}) = \Theta(t_{12})\Theta(t_{23})\ldots\Theta(t_{n-1,n})$. In momentum space

$$\begin{aligned}\tilde\tau(\underline{k}) &= \int \underline{d^2x}e^{ix_ik_i}\tau(\underline{x}) \\ &= \sum_{perm(k)}\int \underline{d^2x}e^{ix_ik_i}\Theta_{1\ldots n}\, w(\underline{x})\,.\end{aligned}$$

The connected Green's functions are given by

$$\tilde\tau(\underline{k}) = \sum_{\underline{k}_1\cup\cdots\cup\underline{k}_m=\underline{k}} \tilde\tau_c(\underline{k}_1)\ldots\tilde\tau_c(\underline{k}_m)\,. \quad (10)$$

For convenience we split off the energy momentum δ-function and define $\tilde\Pi(\underline{k})$by

$$\tilde\tau_c(\underline{k}) = (2\pi)^2\,\delta^{(2)}\left(\textstyle\sum k_i\right)\,\tilde\Xi(\underline{k})\,. \quad (11)$$

S-matrix and form factors. For integrable quantum field theories the n-particle S-matrix factorizes into $n(n-1)/2$ two-particle ones

$$S^{(n)}(\theta_1,\ldots,\theta_n) = \prod_{i<j} S(\theta_{ij})\,,$$

where the product on the right hand side has to be taken in a specific order (see e.g. [17]). The numbers θ_{ij} are the rapidity differences $\theta_{ij} = \theta_i - \theta_j$, which are related to the momenta of the particles by $p_i = m\,(\cosh\theta_i, \sinh\theta_i)$. The form factors of a bosonic field are defined as the matrix elements

$$F(\underline{\theta}) = \langle\, 0\,|\,\varphi(0)\,|\,\theta_1,\ldots,\theta_n\,\rangle \quad (12)$$

(For the paramagnetic phase of the Ising model they are non-zero for $n =$ odd). They satisfy the form factor equations (i) – (v) (see e.g. [22]). We use the normalization $\langle\, 0\,|\,\varphi(0)\,|\,\theta\,\rangle = 1$. As a generalization we write

$$F(\underline{\theta}';\underline{\theta}) = \langle\,\theta'_{n'},\ldots,\theta'_1\,|\,\varphi(0)\,|\,\theta_1,\ldots,\theta_n\,\rangle \quad (13)$$

which is related to (12) by crossing. In particular (see appendix A)

$$F(\theta_1;\theta_2,\theta_3) = F(\theta_1,\theta_2 - i\pi_-,\theta_3 - i\pi_+) + \delta_{\theta_{12}} + \delta_{\theta_{13}} \quad (14)$$

$$F(\theta_2,\theta_3;\theta_4) = F(\theta_3 + i\pi_+,\theta_2 + i\pi_-,\theta_4) + \delta_{\theta_{24}} + \delta_{\theta_{34}} \quad (15)$$

with $i\pi_\pm = i\pi \pm i\epsilon$ and $\delta_{\theta_{12}} = 4\pi\delta(\theta_1 - \theta_2)$.

A. The Green's functions in the low particle approximation

Calculations of the Green's functions of massive theories simplify when we restrict our interest to their imaginary parts, as in the case of Raman scattering (5). In that case for any given energy only limited number of matrix elements contribute to the calculations, namely, those ones which correspond to emissions of particles whose energy does not exceed the threshold.

Below we will derive expressions for the four-point Green's functions in the two-particle approximation.

The 2-point Wightman function in 1-particle intermediate states approximation is (with the short notation $\int_\theta = \frac{1}{4\pi}\int d\theta$)

$$\begin{aligned}w^1(x_1-x_2) &= \int_\theta \langle\, 0\,|\,\varphi(x_1)\,|\,\theta\,\rangle\langle\,\theta\,|\,\varphi(x_2)\,|\,0\rangle \\ &= \int \frac{dp}{2\pi 2\omega}e^{-i(x_1-x_2)p} = i\Delta_+\,(x_1-x_2)\,.\end{aligned}$$

The 4-point Wightman function in 1-0-1 intermediate particle approximation is

$$w^{101}(\underline{x}) = \int_{\theta_1} \langle\, 0\,|\,\varphi(x_1)\,|\,\theta_1\,\rangle\langle\,\theta_1\,|\,\varphi(x_2)\,|\,0\rangle \times \int_{\theta_4} \langle\, 0\,|\,\varphi(x_3)\,|\,\theta_4\rangle\langle\,\theta_4\,|\,\varphi(x_4)\,|\,0\,\rangle = w^1(x_1-x_2)w^1(x_3-x_4)\,. \quad (16)$$

The 4-point Wightman function in 1-2-1 intermediate particle approximation is (with $\int_{\underline{\theta}} = \int_{\theta_1}\dots\int_{\theta_4}$ and $\mathbf{xp} = x_1p_1 + x_2\,(p_2+p_3-p_1) + x_3\,(p_4-p_2-p_3) - x_4p_4$)

$$w^{121}(\underline{x}) = \frac{1}{2}\int_{\underline{\theta}} \langle\, 0\,|\,\varphi(x_1)\,|\,\theta_1\,\rangle\,\langle\,\theta_1\,|\,\varphi(x_2)\,|\,\theta_2,\theta_3\rangle \times \langle\,\theta_3,\theta_2\,|\,\varphi(x_3)\,|\,\theta_4\rangle\,\langle\,\theta_4\,|\,\varphi(x_4)\,|\,0\,\rangle = \frac{1}{2}\int_{\underline{\theta}} e^{-i\mathbf{xp}}F(\theta_1;\theta_2,\theta_3)F(\theta_2,\theta_3;\theta_4)$$

with (see (14) and (15))

$$\begin{aligned} &\tfrac{1}{2}F(\theta_1;\theta_2,\theta_3)F(\theta_2,\theta_3;\theta_4) \\ &= \tfrac{1}{2}\left(F(\theta_1,\theta_2-i\pi_-,\theta_3-i\pi_+) + \delta_{\theta_{12}} + \delta_{\theta_{13}}\right) \\ &\times \left(F(\theta_3+i\pi_+,\theta_2+i\pi_-,\theta_4) + \delta_{\theta_{24}} + \delta_{\theta_{34}}\right) \\ &= I(\underline{\theta}) = I_1(\underline{\theta}) + I_2(\underline{\theta}) + I_3(\underline{\theta})\,. \end{aligned}$$

We have introduced (see appendix A)

$$\begin{aligned} I_1(\underline{\theta}) &= \tfrac{1}{4}F(\theta_1,\theta_2-i\pi_+,\theta_3-i\pi_-) \\ &\quad\times F(\theta_3+i\pi_+,\theta_2+i\pi_-,\theta_4) \\ &\quad+ \tfrac{1}{4}F(\theta_1,\theta_2-i\pi_-,\theta_3-i\pi_+) \\ &\quad\times F(\theta_3+i\pi_-,\theta_2+i\pi_+,\theta_4) \\ I_2(\underline{\theta}) &= \tfrac{1}{4}\left(\delta_{\theta_{12}}\left(1+S(\theta_{23})\right) + \delta_{\theta_{13}}\left(1+S(\theta_{23})\right)\right) \\ &\quad\times F(\theta_3+i\pi_+,\theta_2+i\pi_-,\theta_4) \\ &\quad+ \tfrac{1}{4}F(\theta_1,\theta_2-i\pi_-,\theta_3-i\pi_+) \\ &\quad\times \left(\delta_{\theta_{24}}\left(1+S(\theta_{32})\right) + \delta_{\theta_{34}}\left(1+S(\theta_{32})\right)\right) \\ I_3(\underline{\theta}) &= \tfrac{1}{2}\left(\delta_{\theta_{12}}+\delta_{\theta_{13}}\right)\left(\delta_{\theta_{24}}+\delta_{\theta_{34}}\right)\,. \end{aligned} \quad (17)$$

From I_3 we calculate

$$w_3^{121}(\underline{x}) = w^1\left(x_1-x_4\right)w^1\left(x_2-x_3\right) + w^1\left(x_1-x_3\right)w^1\left(x_2-x_4\right)\,. \quad (18)$$

Therefore neglecting contributions from higher particle intermediate states using (10) and (16) we obtain the connected 4-point Green's function

$$\tilde{\tau}_c(\underline{k}) = \sum_{perm(k)} \int \underline{d^2x}\,\Theta_{1\dots n}e^{ix_ik_i} \times \left(w_1^{121}(\underline{x}) + w_2^{121}(\underline{x})\right) \quad (19)$$

where $w_i^{121}(\underline{x})$ is given by the contribution from $I_i(\underline{\theta})$ in (17). For $k_i = (k_i^0, 0)$ we obtain from (11) (see appendix B)

$$\tilde{\Xi}(\underline{k}) = \frac{1}{32\pi m^6}\sum_{perm(k)} \times \frac{m}{m-k_1^0-i\epsilon}\frac{m}{k_4^0+m-i\epsilon}g\left(\frac{-1}{2m}\left(k_3^0+k_4^0\right)\right) \quad (20)$$

with

$$g(x) = -2\pi\int_\theta \frac{1}{\omega/m}I(0,\theta,-\theta,0)\frac{1}{\omega/m-x} \quad (21)$$

where $I = I_1 + I_2$ contribute. For integrable models typically $S(0) = -1$, then the contribution from I_2 vanishes for $\theta_i \to 0$. With (6) and $G(x) = (x-1)^2\,\mathrm{Im}\,g(x)$ equation (8) follows. Next we consider a simple model, for which we calculate the function $g(x)$ explicitly.

B. The scaling limit of the Ising model

In the scaling limit this model may be described by an interacting Bose field $\sigma_n^z = Cm^{1/8}\phi(x)$, where C is a numerical constant and $m = h - J$. The excitations are noninteracting Majorana fermions with the 2-particle S-matrix $S(\theta) = -1$. The field $\sigma^x = (m/J)^{1/2}\epsilon(x) \sim \bar\psi\psi(x)$, where ψ is a free Majorana spinor field. In [3, 18, 19] the form factor was proposed

$$F(\underline{\theta}) = \langle\,0|\,\sigma(0)\,|\,\theta_1,\dots,\theta_n\,\rangle = (2i)^{\frac{n-1}{2}}\prod_{i<j}\tanh\tfrac{1}{2}\theta_{ij}. \quad (22)$$

For $k_i = (k_i^0, 0)$ in momentum space the contribution from I_2 in (17) vanishes, because $S(0) = -1$. From (17) and (22) we obtain (see appendix B)

$$I_1(0,\theta,-\theta,0) = \tanh^2\theta\coth^4\tfrac{1}{2}\left(\theta-i\epsilon\right) + (\epsilon\to-\epsilon)\,.$$

Substituting it into (21) and taking into account $G(x) = (x-1)^2\,\mathrm{Im}\,g(x)$ and the relation between σ^z and ϕ we obtain (9).

From $\epsilon(x) \sim \bar\psi\psi(x)$ one has for a free Majorana spinor field

$$\langle 0|\epsilon(0)|\theta_1,\theta_2\rangle = \sinh(\theta_{12}/2). \quad (23)$$

For low intermediate particle numbers this leads to (4) as above (see appendix C).

IV. CONCLUSIONS AND ACKNOWLEDGEMENTS

We calculated the three and the four point correlation functions for the ferromagnetic Quantum Ising model and discussed their relation to the observable quantities. In the paramagnetic phase of FQI the magnetic field is directly coupled to the spin operator which has matrix elements between states with odd and even number of the

Ising fermions. The fact that light can create odd number of fermionic excitations is quite remarkable. It emphasizes an ambiguity between bosons and fermions existing in one dimension.

As we have said in Introduction, the best experimental realization of FQI model known to date is found in columbite $CoNb_2O_6$ [6],[7],[8]. Another possible candidate is Sr_3CuIrO_6 [16]. Both these materials are quasi 1D insulators; the columbite displays a quantum critical point at $B = 5.5$T which is very well described by the theory of the Ising model [7]. Neutron scattering [6] and terahertz spectroscopy [8] also yield excellent agreement with the theoretical predictions. In the view of these we suggest that a good test of our theory would be high field spectroscopic measurements at terahertz frequencies on $CoNb_2O_6$.

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Appendix A: Crossing

The form factors (13) satisfy crossing relations (see e.g. (31) in [21]), in particular

$$\begin{aligned} F(\theta_1;\theta_2,\theta_3) &= \langle\,\theta_1\,|\,\varphi(0)\,|\,\theta_2,\theta_3\rangle = F(\theta_1+i\pi_-,\theta_2,\theta_3)+\delta_{\theta_{12}}+\delta_{\theta_{13}}S(\theta_{23}) \\ F(\theta_2,\theta_3;\theta_4) &= \langle\,\theta_3,\theta_2\,|\,\varphi(0)\,|\,\theta_4\rangle = F(\theta_3+i\pi_-,\theta_2+i\pi_-,\theta_4)+\delta_{\theta_{24}}+\delta_{\theta_{34}}S(\theta_{32}) \end{aligned} \tag{A.1}$$

with $i\pi_\pm = i\pi \pm i\epsilon$ and $\delta_{\theta_{12}} = 4\pi\delta(\theta_1-\theta_2)$. Using the form factor equation (iii) and Lorentz invariance (see e.g. [22])

$$\operatorname*{Res}_{\theta_{12}=i\pi} F(\theta_1,\theta_2,\theta_3) = 2i\,(\mathbf{1}-S(\theta_{23}))$$

$$F(\theta_1,\theta_2,\theta_3) = F(\theta_1+\mu,\theta_2+\mu,\theta_3+\mu)$$

we can rewrite these equations as (14) and (15). And further one derives

$$\tfrac12 F(\theta_1,\theta_2-i\pi_-,\theta_3-i\pi_+)+\delta_{\theta_{12}}+\delta_{\theta_{13}} = \tfrac12\left(F(\theta_1,\theta_2-i\pi_+,\theta_3-i\pi_-)+\delta_{\theta_{12}}\left(1+S(\theta_{23})\right)+\delta_{\theta_{13}}\left(1+S(\theta_{23})\right)\right) \tag{A.2}$$

$$\tfrac12 F(\theta_3+i\pi_+,\theta_2+i\pi_-,\theta_4)+\delta_{\theta_{24}}+\delta_{\theta_{34}} = \tfrac12\left(F(\theta_3+i\pi_-,\theta_2+i\pi_+,\theta_4)+\delta_{\theta_{24}}\left(1+S(\theta_{32})\right)+\delta_{\theta_{34}}\left(1+S(\theta_{32})\right)\right)\,. \tag{A.3}$$

Using equations (14,15) and the identity
$(a+b+c)\,(d+e+f) = \left(\tfrac12 a+b+c\right)d + a\left(\tfrac12 d+e+f\right)+(b+c)\,(e+f)$ we derive

$$\begin{aligned} &F(\theta_1;\theta_2,\theta_3)F(\theta_2,\theta_3;\theta_4) \\ &= \left(F(\theta_1,\theta_2-i\pi_-,\theta_3-i\pi_+)+\delta_{\theta_{12}}+\delta_{\theta_{13}}\right)\left(F(\theta_3+i\pi_+,\theta_2+i\pi_-,\theta_4)+\delta_{\theta_{24}}+\delta_{\theta_{34}}\right) \\ &= \left(\tfrac12 F(\theta_1,\theta_2-i\pi_-,\theta_3-i\pi_+)+\delta_{\theta_{12}}+\delta_{\theta_{13}}\right)F(\theta_3+i\pi_+,\theta_2+i\pi_-,\theta_4) \\ &+ F(\theta_1,\theta_2-i\pi_-,\theta_3-i\pi_+)\left(\tfrac12 F(\theta_3+i\pi_+,\theta_2+i\pi_-,\theta_4)+\delta_{\theta_{24}}+\delta_{\theta_{34}}\right) \\ &+ \left(\delta_{\theta_1\theta_2}+\delta_{\theta_1\theta_3}\right)\left(\delta_{\theta_4\theta_2}+\delta_{\theta_4\theta_3}\right) \end{aligned}$$

then (A.2) and (A.3) prove (17).

Appendix B: Proof of (8)

1. Calculation of τ_c^{121}:

To derive (20) from (19) we calculate (for $i=1,2$)

$$\begin{aligned} &\int \underline{d^2x}\,\Theta_{1\dots n}e^{ix_ik_i}\int_{\underline{p}} e^{-ix_1p_1-ix_2(p_2+p_3-p_1)-ix_3(p_4-p_2-p_3)+ix_4p_4}I_i(\theta_1,\theta_2,\theta_3,\theta_4) \\ &= \int \underline{dx}^0\,\Theta_{1\dots n}e^{ix_i^0k_i^0}\int_{\underline{\theta}} e^{-ix_1^0\omega_1-ix_2^0(\omega_2+\omega_3-\omega_1)-ix_3^0(\omega_4-\omega_2-\omega_3)+ix_4^0\omega_4} \\ &(2\pi)^4\,\delta(p_1-k_1^1)\delta(p_2+p_3-p_1-k_2^1)\delta(p_4-p_2-p_3-k_3^1)\delta(p_4+k_4^1)I_i(\theta_1,\theta_2,\theta_3,\theta_4). \end{aligned}$$

For $k_i = (k_i^0,0)$ this is equal to

$$\begin{aligned} &= 2\pi\delta\left(k_1^1+k_2^1+k_3^1+k_4^1\right)\int \underline{dx}^0\,\Theta_{1\dots n}e^{ix_i^0k_i^0}\int_{\underline{\theta}} e^{-ix_1^0m-ix_2^0(\omega_2+\omega_3-m)-ix_3^0(m-\omega_2-\omega_3)+ix_4^0m} \\ &\times (2\pi)^3\,\delta(p_1)\delta(p_2+p_3)\delta(p_4)I_i(0,\theta_2,-\theta_2,0) \\ &= 2\pi\delta\left(k_1^1+k_2^1+k_3^1+k_4^1\right)\frac{1}{(2m)^2}\int_\theta \frac{1}{2\omega}I_i(0,\theta,-\theta,0)\int_{-\infty}^{\infty}dx_1^0\int_{-\infty}^{0}dx_2^0\int_{-\infty}^{0}dx_3^0\int_{-\infty}^{0}dx_4^0 \\ &\times e^{ix_1^0\left(k_1^0-m\right)+i\left(x_2^0+x_1^0\right)\left(k_2^0-(2\omega-m)\right)+i\left(x_3^0+x_2^0+x_1^0\right)\left(k_3^0-(m-2\omega)\right)+i\left(x_4^0+x_3^0+x_2^0+x_1^0\right)\left(k_4^0+m\right)} \end{aligned}$$

and

$$\begin{aligned} &\int_{-\infty}^{\infty}dx_1^0\int_{-\infty}^{0}dx_2^0\int_{-\infty}^{0}dx_3^0\int_{-\infty}^{0}dx_4^0 e^{ix_1^0\left(k_1^0-m\right)+i\left(x_2^0+x_1^0\right)\left(k_2^0-(2\omega-m)\right)+i\left(x_3^0+x_2^0+x_1^0\right)\left(k_3^0-(m-2\omega)\right)+i\left(x_4^0+x_3^0+x_2^0+x_1^0\right)\left(k_4^0+m\right)} \\ &= 2\pi\delta\left(k_1^0+k_2^0+k_3^0+k_4^0\right)\frac{-i}{k_2^0+k_3^0+k_4^0+m-i\epsilon}\,\frac{-i}{k_3^0+k_4^0+2\omega-i\epsilon}\,\frac{-i}{k_4^0+m-i\epsilon} \end{aligned}$$

proves (20) and (21). For integrable models typically $S(0)=-1$, then the contribution from I_2 vanishes for $\theta_i \to 0$ (for the scaling Ising model we have $S(\theta)\equiv -1$).

2. The function $g(x)$ for the scaling Ising model:

From (17) and (22) we obtain (up to const)

$$\begin{aligned} I_1(0,\theta,-\theta,0) &= \tfrac{1}{4}F(0,\theta-i\pi_+,-\theta-i\pi_-)F(-\theta+i\pi_+,\theta+i\pi_-,0)+(\epsilon\to-\epsilon) \\ &= \left(\left(\tanh\frac{1}{2}\left(-\theta+i\pi+i\epsilon\right)\right)\left(\tanh\frac{1}{2}\left(\theta+i\pi-i\epsilon\right)\right)\tanh\frac{1}{2}\left(2\theta\right)\right) \\ &\times\left(\left(\tanh\frac{1}{2}\left(-2\theta\right)\right)\left(\tanh\frac{1}{2}\left(-\theta+i\pi+i\epsilon\right)\right)\tanh\frac{1}{2}\left(\theta+i\pi-i\epsilon\right)\right)+(\epsilon\to-\epsilon) \\ &= \tanh^2\theta\coth^4\tfrac{1}{2}\left(\theta-i\epsilon\right)+(\epsilon\to-\epsilon)\,. \end{aligned}$$

and

$$\begin{aligned} g(x) &= -2\pi\int_\theta \frac{1}{\omega/m} I_1(0,\theta,-\theta,0)\frac{1}{\omega/m-x} = -\frac{1}{2}\int_{-\infty}^{\infty} d\theta\left(\frac{\coth^4\frac{1}{2}\left(\theta-i\epsilon\right)\tanh^2\theta+(\epsilon\to-\epsilon)}{\cosh\theta\left(\cosh\theta-x\right)}\right) \\ &= \frac{16}{1-x}-\frac{15\pi}{2x}-\frac{8}{x}-\frac{4\pi+2}{x^2}-\frac{\pi}{x^3}-\frac{(x+1)^2\sqrt{x^2-1}}{x^3\left(x-1\right)^2}2\ln\left(-x+\sqrt{x^2-1}\right) \end{aligned} \qquad \text{(B.1)}$$

with $g(0)=10\pi+\frac{94}{3}$ (see also [20]) and the imaginary part for $x>1$

$$\operatorname{Im} g(x\pm i\epsilon) = \pm\Theta(x-1)2\pi\frac{(x+1)^2\sqrt{x^2-1}}{x^3\left(x-1\right)^2}\,. \qquad \text{(B.2)}$$

3. The 4-point Ξ-function and calculation of $\chi(\omega_i,\omega_f)$:

The sum over all permutations in (20) yields

$$\begin{aligned} \Xi(\underline{k}) = \frac{1}{32\pi m^6}\Bigg\{ &\left(\frac{m}{k_4^0+m}+\frac{m}{k_3^0+m}\right)\left(\frac{m}{-k_1^0+m}+\frac{m}{-k_2^0+m}\right)g\left(\frac{-1}{2m}\left(k_3^0+k_4^0\right)\right) \\ +&\left(\frac{m}{k_4^0+m}+\frac{m}{k_2^0+m}\right)\left(\frac{m}{-k_1^0+m}+\frac{m}{-k_3^0+m}\right)g\left(\frac{-1}{2m}\left(k_2^0+k_4^0\right)\right) \\ +&\left(\frac{m}{k_4^0+m}+\frac{m}{k_1^0+m}\right)\left(\frac{m}{-k_2^0+m}+\frac{m}{-k_3^0+m}\right)g\left(\frac{-1}{2m}\left(k_1^0+k_4^0\right)\right) \\ +&\left(\frac{m}{k_3^0+m}+\frac{m}{k_1^0+m}\right)\left(\frac{m}{-k_2^0+m}+\frac{m}{-k_4^0+m}\right)g\left(\frac{-1}{2m}\left(k_1^0+k_3^0\right)\right) \\ +&\left(\frac{m}{k_2^0+m}+\frac{m}{k_1^0+m}\right)\left(\frac{m}{-k_3^0+m}+\frac{m}{-k_4^0+m}\right)g\left(\frac{-1}{2m}\left(k_1^0+k_2^0\right)\right) \\ +&\left(\frac{m}{k_2^0+m}+\frac{m}{k_3^0+m}\right)\left(\frac{m}{-k_1^0+m}+\frac{m}{-k_4^0+m}\right)g\left(\frac{-1}{2m}\left(k_3^0+k_2^0\right)\right)\Bigg\} \end{aligned}$$

Substituting this into (6) we obtain

$$\begin{aligned} \chi(\omega_i,\omega_f) \sim &\frac{(\omega_i-\omega_f+2m)^2\operatorname{Im} g\left(\frac{-1}{2m}\left(\omega_i-\omega_f-i\delta_{12}\right)\right)}{(\omega_i+m)^2\left(\omega_f-m\right)^2}+\frac{(\omega_i+\omega_f+2m)^2\operatorname{Im} g\left(\frac{-1}{2m}\left(\omega_i+\omega_f-i\delta_{12}\right)\right)}{(\omega_i+m)^2\left(\omega_f+m\right)^2} \\ +&\frac{(\omega_i+\omega_f-2m)^2\operatorname{Im} g\left(\frac{1}{2m}\left(\omega_i+\omega_f+i\delta_{12}\right)\right)}{(\omega_f-m)^2\left(\omega_i-m\right)^2}+\frac{(\omega_i-\omega_f-2m)^2\operatorname{Im} g\left(\frac{1}{2m}\left(\omega_i-\omega_f+i\delta_{12}\right)\right)}{(\omega_f+m)^2\left(\omega_i-m\right)^2} \end{aligned}$$

At $\omega_i>\omega_f>0$ only the last two terms remain and (8) follows with $G(x)=(x-1)^2\operatorname{Im} g(x)$.

Appendix C: Proof of (4):

We consider the 3 point Greens function

$$\tau_{\varphi\varphi\epsilon}(\underline{x}) = \langle\, 0\,|\, T\varphi(x_1)\,\varphi(x_2)\epsilon(x_3)|\,0\,\rangle$$

and the Fourier transform (for $\varphi_1,\varphi_2,\varphi_3 = \varphi,\varphi,\epsilon$)

$$\begin{aligned}\tilde{\tau}_{\varphi\varphi\epsilon}(\underline{k}) &= \int \underline{d^2x} e^{ix_ik_i}\tau_{\varphi_1\varphi_2\varphi_3}(\underline{x}) = \sum_{\pi\in S_3}\int \underline{d^2x} e^{ix_i\pi k_i}\Theta_{123}\,\langle\varphi_{\pi1}(x_1)\,\varphi_{\pi2}(x_2)\varphi_{\pi3}(x_3)\rangle \\ &= (2\pi)^2\,\delta^{(2)}\,(k_1+k_2+k_3)\,\tilde{\Xi}_{\varphi\varphi\epsilon}(\underline{k}).\end{aligned} \tag{C.1}$$

The 3-point Wightman functions in low intermediate particle number approximation are

$$\begin{aligned} w^{12}_{\varphi\varphi\epsilon}(\underline{x}) &= \frac{1}{2!}\int_{\underline{\theta}}\langle\, 0\,|\,\varphi(x_1)\,|\,\theta_1\,\rangle\langle\,\theta_1\,|\,\varphi(x_2)\,|\,\theta_2,\theta_3\rangle\langle\,\theta_3,\theta_2\,|\,\epsilon(x_3)\,|\,0\,\rangle \\ w^{11}_{\varphi\epsilon\varphi}(\underline{x}) &= \int_{\underline{\theta}}\langle\, 0\,|\,\varphi(x_1)\,|\,\theta_1\,\rangle\;\langle\,\theta_1\,|\,\epsilon(x_2)\,|\,\theta_2\rangle\,\langle\,\theta_2\,|\,\varphi(x_3)\,|\,0\,\rangle \\ w^{21}_{\epsilon\varphi\varphi}(\underline{x}) &= \frac{1}{2!}\int_{\underline{\theta}}\langle\, 0\,|\,\epsilon(x_1)\,|\,\theta_1,\theta_2\,\rangle\,\langle\,\theta_1\,,\theta_2|\,\varphi(x_2)\,|\,\theta_3\rangle\,\langle\theta_3\,|\,\varphi(x_3)\,|\,0\,\rangle. \end{aligned}$$

As above using the form factor formulas (23), (22) and the crossing relations (A.1) one obtains the Fourier transforms (for $k_i = (k_i^0, 0)$)

$$\begin{aligned} \tilde{\Xi}^{12}_{\varphi\varphi\epsilon}(k_1,k_2,k_3) &= \frac{-i}{32\pi m^4}\frac{m}{k_1^0-m+i\epsilon}h\left(-k_3^0/(2m)+i\epsilon\right) \\ \tilde{\Xi}^{11}_{\varphi\epsilon\varphi}(k_1,k_2,k_3) &= -\frac{1}{4}\frac{i}{m^4}\frac{m}{m-k_1^0-i\epsilon}\frac{m}{m+k_3^0-i\epsilon} \\ \tilde{\Xi}^{21}_{\epsilon\varphi\varphi}(k_1,k_2,k_3) &= \frac{-i}{32\pi m^4}\frac{m}{-k_3^0-m+i\epsilon}h\left(k_1^0/(2m)+i\epsilon\right) \end{aligned}$$

with

$$h(x) = \int_{-\infty}^{\infty}\left(1+\frac{1}{\cosh\theta}\right)^2\frac{1}{\cosh\theta - x}d\theta = -\frac{2}{x}\pi - \frac{2}{x} - \frac{1}{x^2}\pi - 2\frac{x^2+2x+1}{x^2\sqrt{x^2-1}}\ln\left(-x-\sqrt{x^2-1}\right).$$

Finally with (C.1) we obtain

$$\tilde{\Xi}_{\varphi\varphi\epsilon}(k_1,k_2,k_3) = \tilde{\Xi}^{12}_{\varphi\varphi\epsilon}(k_1,k_2,k_3) + \tilde{\Xi}^{11}_{\varphi\epsilon\varphi}(k_1,k_3,k_2) + \tilde{\Xi}^{21}_{\epsilon\varphi\varphi}(k_3,k_1,k_2) + (k_1\leftrightarrow k_2)$$

which proves (4).