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Finite-temperature lineshapes in gapped quantum spin chains

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We consider the finite-temperature dynamical susceptibility $\chi(\omega,q,T)$ of gapped quantum spin chains such as the integer spin Heisenberg and transverse field Ising models. At zero temperature χ in these models is dominated by a delta-function line arising from the coherent propagation of single-particle modes. Using methods of integrable quantum field theory, we determine the evolution of the lineshape at low temperatures. We show that the lineshape is in general asymmetric in energy. We discuss the application of our results to inelastic neutron-scattering experiments on gapped spin chain systems such as Y_2BaNiO_5 and $CsNiCl_3$.

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Quasi-one-dimensional spin chains are materials where quantum fluctuations give rise to striking strongly correlated phenomena. An example of such behavior is the distinction, first identified by Haldane,[1] between integer and half-integer Heisenberg spin chains. The former are gapped while the latter are gapless. The dynamics of such spin chains can be probed by inelastic neutron scattering, which makes it possible to determine with impressive accuracy the spectrum of spin excitations together with their lifetimes.[2]

The zero-temperature properties of Haldane-gap chains are thus now well understood:[3] the dynamical susceptibility (DS) is dominated by a coherent triplet of magnon modes above a gap Δ in the vicinity of the antiferromagnetic wave number π/a_0 (a_0 is the lattice spacing). At energies above 3Δ there is a weak three magnon scattering continuum.[4] The coherent magnon triplet disperses upwards in energy when the momentum is decreased from π/a_0 and at small momenta disappears in a weak two magnon scattering continuum.[5] Recently the crossover in the spin dynamics from the strongly correlated $T=0$ quantum regime to the classical high-temperature regime has started to be addressed.[6–9] In a system that supports a coherent, gapped, magnetic single-particle excitation at $T=0$, the question then arises of how the corresponding delta function in the DS broadens at finite temperatures.[7,10–12] A partial answer to this question has been given in Refs. 10 and 11 where it was shown that at $T\ll\Delta$ the broadening in the immediate vicinity of the gap is Lorentzian in form.

In the present work we develop an approach to computing finite-temperature DSs and use it to determine the *entire* lineshape. As our central finding, we demonstrate that the lineshape is markedly asymmetric in energy, a feature that becomes more pronounced as the temperature increases. While we focus upon the lineshape in gapped quantum spin chains, we stress that this approach is applicable to the calculation of general response functions in generic integrable continuum models such as those considered in Refs. 13–16.

General theoretical framework. The systems we consider here all have representations as general Heisenberg models:

$$H=\sum_i J_\perp \mathbf{S}_{\perp i}\cdot\mathbf{S}_{\perp i+1}+J_z S_{zi}S_{zi+1}+\mathbf{H}\cdot\mathbf{S}. \tag{1}$$

Here $\mathbf{S}_i=(\mathbf{S}_{\perp i},S_{zi})$ is a quantum spin (either integer or half-integer) at chain site i. We allow the spin chain to have anisotropic couplings $(J_\perp,J_z)$ and for a magnetic field $\mathbf{H}$ to be present. We are interested in computing the dynamical susceptibility

$$\chi(\omega,q)=-\int_0^\beta d\tau\int dx e^{i\omega_n\tau-iqx}\langle\mathbf{S}(\tau,x)\mathbf{S}(0)\rangle|_{\omega_n\to-i\omega+\delta}. \tag{2}$$

To that end we expand $C(\tau,x)\equiv\langle\mathbf{S}(\tau,x)\mathbf{S}(0)\rangle$ in a basis $\{|l\rangle\}$ of exact eigenstates of H,

$$C(\tau,x)=\frac{1}{\mathcal{Z}}\sum_{l,m}e^{-\beta E_l}\langle l|S_z(\tau,x)|m\rangle\langle m|S_z(0)|l\rangle, \tag{3}$$

where E_l is the energy of eigenstate $|l\rangle$ and $\mathcal{Z}=\sum_l e^{-\beta E_l}$ is the partition function of the theory. Due to the gap Δ in this system, the Fourier transform of $C(\tau,x)$ has a well-defined low-temperature expansion.

This representation of $\chi(\omega,q)$ finds its virtue when we take the continuum limit of the lattice model in Eq. (1). In such cases the matrix elements $\langle l|S_z(0)|m\rangle$ can be determined exactly. At $T=0$ this permits the exact computation of $\mathrm{Im}\,\chi(\omega,q)$ at energies up several times the gap through the computation of a small number of matrix elements. At finite temperatures, this approach, for the problem at hand, breaks down in two ways: (i) the needed matrix elements (as well as $\mathcal{Z}$) become highly singular objects; and (ii) to obtain the finite-temperature broadening of the coherent mode, an infinite number of matrix elements are needed. We solve these problems in a two-step fashion. The singularities of the matrix elements are a direct consequence of working in the infinite volume, where eigenstates are normalized to delta functions. While it is possible in certain circumstances to deal with these singularities directly,[13–15,17,18] to circumvent this first difficulty we instead work with chains of large but finite length R. The infinities in the matrix elements are then reduced to terms merely proportional to R which are cleanly cancelled by similar terms in the partition function. Finally, we take the limit $R\to\infty$ of the resulting expressions, thus eliminating any finite-size effects. To handle the second difficulty, we recognize that the *infinite* subset of needed matrix elements from the sum in Eq. (3) can be organized according to a Dyson's equation. This allows us to characterize the subset by resumming a *finite* number of matrix elements, resulting in a low-T expansion where $\exp(-\Delta/T)$ is the small parameter. We now apply this approach to two experimentally relevant cases: the transverse field Ising model (TFIM) and the spin-1 chain as represented by the O(3) nonlinear sigma model.

Transverse field Ising model. The ferromagnetic TFIM is obtained by taking spins 1/2 in Eq. (1) and setting $J_\perp=0$, $\mathbf{H}=-h\hat{x}$, and $J^z=-2J<0$. In the vicinity of the critical point $J=h$, this theory has a continuum representation as a free Majorana fermion:

$$H=\frac{1}{2\pi}\int_0^R dx\frac{iv}{2}(\bar{\psi}\partial_x\bar{\psi}-\psi\partial_x\psi)-i\Delta\psi\bar{\psi}. \quad (4)$$

Here $\psi(x,t)$ and $\bar{\psi}(x,t)$ are the right and left components of a Majorana Fermi field. The gap of the fermions in the disordered regime $J<h$ is given by $\Delta=|J-h|$. The Hilbert space of the theory on a ring of circumference R divides itself into so-called Neveu-Schwarz (NS) and Ramond (R) sectors. The NS sector consists of a Fock space built with even numbers of half-integer fermionic modes, i.e., states of the form $|p_1\cdots p_{2N}\rangle_{\rm NS}\equiv a^\dagger_{p_1}\cdots a^\dagger_{p_{2N}}|0\rangle_{\rm NS}$ where $p_i=2\pi(n_i+1/2)/R$, with n_i an integer, while the R sector consists of a Fock space composed of odd numbers of even integer fermionic modes $|k_1\cdots k_{2M+1}\rangle_R\equiv\{a^\dagger_{k_1}\cdots a^\dagger_{k_{2M+1}}|0\rangle_R\}$, $k_i=2\pi n_i/R$. Momentum and energy of a NS state $|p_1\cdots p_{2N}\rangle_{\rm NS}$ are given simply by $P(p_i)=\Sigma_i^{2N}p_i$ and $E(p_i)=\Sigma_i^{2N}\epsilon(p_i)$, respectively, where $\epsilon(p)=\sqrt{p^2+\Delta^2}$. An analogous relation holds for states in the R sector. To compute the DS of this model, we need access to the matrix elements $\langle l|S_z|m\rangle$ of the spin operator S_z at large, finite R. These matrix elements, derived in Refs. 19 and 20, only are nonzero when $|l\rangle$ and $|m\rangle$ belong to different sectors, in which case

$$\begin{aligned}&{}_R\langle k_1\cdots k_{2M+1}|S_z(0)|p_1\cdots p_{2N}\rangle_{\rm NS}\\&\quad=C_R\prod_{i,j}g(\theta_{k_i})g(\theta_{p_j})\\&\qquad\times\prod_{i<j}f(\theta_{k_i}-\theta_{k_j})\prod_{i<j}f(\theta_{p_i}-\theta_{p_j})\prod_{i,j}f^{-1}(\theta_{k_i}-\theta_{p_j}).\end{aligned} \quad (5)$$

Here $C_R=C^{1/2}+\mathcal{O}(e^{-\Delta R})$ is a known normalization, θ_{p_i} parameterizes the momentum p_i via $p_i=\Delta\sinh(\theta_{p_i})$, $g(\theta)=-i[\Delta R\cosh(\theta)]^{-1/2}$, and $f(\theta)=\tanh(\theta/2)$. Note that up to exponentially small corrections, these matrix elements have the same form as at $R=\infty$. The sole difference in the two cases is that at finite R, the momenta are quantized. This is a pattern that repeats itself for general integrable models, as emphasized in Ref. 21, and that we will exploit for our analysis of spin-1 chains.

Crucially all matrix elements (5) are finite, a consequence of working at finite R. The sole possible divergences come from the term $\Pi_{i,j}f^{-1}(\theta_{k_i}-\theta_{p_j})$ and occur as two momenta, k_i and p_j, approach one another. However, as k_i lies in the R sector with integer quantization and p_j lies in the NS sector with half-integer quantization, the two are never exactly equal provided R is finite. In contrast, at $R=\infty$ the distinction between the R and NS sectors collapses (via a spontaneous Z_2 symmetry breaking). Concomitantly, S_z has matrix elements where k_i and p_j may, in principle, be equal, and so which are infinite. By working at large, finite R, we thus obtain a clean, unambiguous regulation of these infinities.

Even though any given matrix element is finite, we must still sum an infinite number of matrix elements in the Lehmann expansion of Eq. (3) in order to obtain the DS χ^I at finite T. To do so we employ a Dyson-like equation by writing $\chi^I(\omega,q)$ in the form

$$\chi^I(\omega,q)=D^I(\omega,q)/[1-D^I(\omega,q)\Sigma^I(\omega,q)]. \quad (6)$$

Here $D^I(\omega,q)=2C/[(\omega+i\delta)^2-\epsilon^2(q)]$ is the DS in the absence of temperature induced interactions. We emphasize that we only consider frequencies close to Δ and low temperatures so that we can neglect n-particle contributions to D with $n\geq 3$ as their contributions are vanishingly small. As $\chi^I(\omega,q)$ has a well-defined low-temperature expansion, so must $\Sigma^I(\omega,q)$: $\Sigma^I(\omega,q)=\Sigma_n\Sigma^I_n(\omega,q)$, where Σ^I_n is at best $\mathcal{O}(e^{-n\beta\Delta})$. We can readily compute Σ^I_1. To do so we expand $\chi^I=D^I+(D^I)^2\Sigma^I_1+\mathcal{O}(e^{-2\beta\Delta})$ and then compare with the Lehmann expansion of Eq. (3). To facilitate this we divide $C^I(x,\tau)$ into contributions coming from matrix elements with a fixed number of excitations on either side of the operator S_z, i.e., $C^I(x,\tau)=\Sigma_{M,N}C^I_{M,N}(x,\tau)$ where

$$\begin{aligned}C^I_{M,N}(x,\tau)=&\sum_{(k_j),(p_i)}|\langle k_1\cdots k_M|S_z(0)|p_1\cdots p_N\rangle|^2\\&\times e^{-\beta E(k_j)}e^{-\tau[E(p_i)-E(k_j)]+ix[P(p_i)-P(k_j)]}.\end{aligned} \quad (7)$$

Kinematic constraints give that $C^I_{M,N}(\omega,q)$ is of order $e^{-\beta\max[N\Delta-\theta(\omega)\omega,M\Delta+\theta(-\omega)\omega]}$. Keeping terms to at least $\mathcal{O}(e^{-\beta\Delta})$ and such that $\mathrm{Im}\,\chi^I(-\omega,-q)=-\mathrm{Im}\,\chi^I(\omega,q)$, we reduce $\chi^I(\omega,q)$ to $\chi^I=\frac{1}{\mathcal{Z}}(C^I_{01}+C^I_{10}+C^I_{12}+C^I_{21}+e^{-2\beta\epsilon(q)}D^I)$. The final term $e^{-2\beta\epsilon(q)}D^I$ is a "disconnected" contribution arising from $C_{23}+C_{32}$, exactly cancelling off a similar contribution appearing in $C_{21}+C_{12}$. Comparing these two expansions for χ^I gives us an expression for Σ_1. First expanding out the partition function $\mathcal{Z}^I=\Sigma^\infty_{n=0}Z^I_n$ where $Z^I_0=1$, $Z^I_1=\Sigma_{p\in R}e^{-\beta\epsilon(p)}$, and generally Z^I_n is $\mathcal{O}(e^{-n\beta\Delta})$, and then noting that $C^I_{01}+C^I_{10}=(1-e^{-\beta\epsilon(q)})D^I$, we obtain for Σ^I_1, $\Sigma^I_1=(C^I_{12}+C^I_{21})(D^I)^{-2}-(Z^I_1+e^{-\beta\epsilon(q)})(1-e^{-\beta\epsilon(q)})(D^I)^{-1}$. In the limit $R\to\infty$ we obtain a representation for Σ^I_1 in terms of a finite integral.

In Fig. 1 we plot the resulting $\mathrm{Im}\,\chi^I(\omega,q=0)$ at a variety of temperatures. At $T=0.25\Delta$, the DS is approximately Lorentzian, but as the temperature is increased to $T=0.6\Delta$, the lineshape develops a marked asymmetry. This asymmetry is implicit (though not specifically discussed) in a virial-like expansion of the finite T DS.[22] We quantify the amount of asymmetry in the lineshape by computing the ratio $A_{\rm LR}$ of the spectral weight to the left and the right of $\omega=\Delta$. In the inset of Fig. 1 we compare our results for $T=0.5\Delta$ (black solid curve) to those arrived at by using a semiclassical approach[10] (blue curve) and to a Lorentzian fit thereof (red dashed curve). We see that our computation yields comparatively far stronger asymmetries in the lineshape. While the semiclassic has a slight asymmetry at $T=0.5\Delta$, it is close to being Lorentzian.

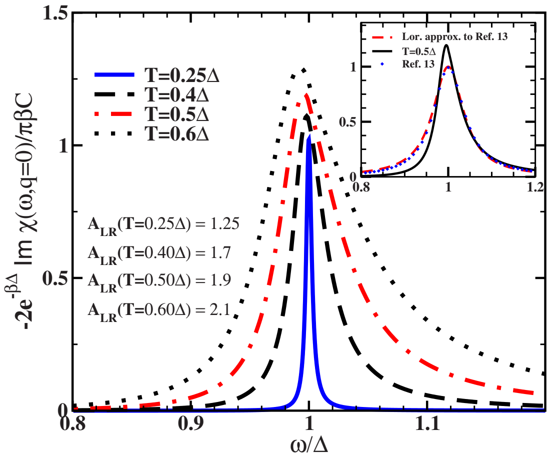

FIG. 1. (Color online) Imaginary part of $\chi(\omega,q)$ for several temperatures for the TFIM in the thermodynamic limit.

The asymmetry of the lineshape can be understood qualitatively as a product of two factors. The first factor is the joint density of states that arises in Born-like approximation for a neutron experiencing energy loss ω in scattering off a thermally excited magnon with energy $E_i=\epsilon(p_1)$ and momentum $P_i=p_1$ into an unoccupied two magnon state with energy $E_f=\epsilon(p_2)+\epsilon(p_3)$ and momentum $P_f=p_2+p_3$:

$$D(\omega,P) \propto \sum_{\{p_j\}} n(p_1)\bar{n}_{23}\delta_{P+P_i,P_f}\delta_{\omega+E_i,E_f}. \quad (8)$$

Here $n(p)=[1+\exp(\beta\epsilon(p)\ ]^{-1}$ and $\bar{n}_{23}=[1-n(p_2)][1-n(p_3)]$ are the thermal occupation numbers for free fermions. This density of states $D(\omega,P)$ is skewed toward higher frequencies. The second factor is the momentum dependence of the matrix elements governing this scattering process [see Eq. (5)]. This latter effect is dispositive as it is absent in the semiclassics.

Spin-1 Heisenberg model. We now apply our approach to the thermal broadening of the coherent mode in a gapped isotropic spin-1 chain [i.e., taking spin $S=1$, $J_\perp=J_z\equiv J$, and $H=0$ in Eq. (1)]. The continuum limit of the isotropic spin-1 chain is given by the O(3) nonlinear sigma model with Lagrangian[1] $L=\frac{1}{2g}\int dx[(\partial_t\mathbf{n}\cdot\partial_t\mathbf{n}-\partial_x\mathbf{n}\cdot\partial_x\mathbf{n})]$. The lattice spin operators S_i are related to the continuum fields by $\mathbf{S}_j\simeq(-1)^j\mathbf{n}(ja_0)+\frac{1}{g}\mathbf{n}\times\partial_t\mathbf{n}$ (with a_0 the lattice spacing).[23] In this Rapid Communication we will focus on the DS near wave vector $q=\pi/a_0$ and so be interested in computing $C(x,\tau)=\langle n^z(x,\tau)n(0)\rangle$. The spectrum and scattering matrix of the O(3) nonlinear sigma model (NLSM) are known exactly. There are three elementary excitations, $A_a^\dagger(\theta)$, $a=x,y,z$, forming a vector representation of O(3). The excitations have a gap behaving as $\Delta\sim Je^{-1/g}$. For $S=1$ the gap is an appreciable fraction of Δ thus limiting the applicability of our results to energies ω not far different than Δ. The excitations' energy and momentum are parametrized in terms of the rapidity θ via $\epsilon(\theta)=\Delta\cosh(\theta)$ and $p(\theta)=\Delta\sinh(\theta)$.

Like the TFIM, the eigenstates of the O(3) NLSM can be delineated exhaustively in terms of scattering states, i.e., $|\theta_1,a_1;\cdots,\theta_n,a_n\rangle=A_{a_1}^\dagger(\theta_1)\cdots A_{a_N}^\dagger(\theta_N)|0\rangle$. However, the matrix elements of these states involving the operator n_z are considerably more complicated than those of the TFIM. However, as we are working at low temperatures, to compute the DS, χ^{O_3}, we will only need recourse to matrix elements involving a maximum of three excitations (as with the TFIM). In infinite volume they are given by:[17,24]

$$\langle\theta_1,a_1|n^a(0)|\theta_3,a_3;\theta_2,a_2\rangle$$
$$=-\sqrt{C}\frac{\pi^3}{2}\psi(\hat{\theta}_{21})\psi(\hat{\theta}_{31})$$
$$\times\psi(\theta_{23})[\delta_{aa_1}\delta_{a_2a_3}\theta_{23}+\delta_{aa_2}\delta_{a_1a_3}\hat{\theta}_{31}+\delta_{aa_3}\delta_{a_1a_2}\hat{\theta}_{12}],$$
$$\langle 0|n^a(0)|\theta,b\rangle=\sqrt{C}\delta_{ab}, \quad (9)$$

where C is a normalization, $\psi(\theta)=\frac{\theta+i\pi}{\theta(2\pi i+\theta)}\tanh^2(\theta/2)$, $\hat{\theta}=\theta-i\pi$, and $\theta_{12}=\theta_1-\theta_2$. As with the TFIM model, we work in a large finite volume. The sole effect of doing so upon the matrix elements, up to negligible $e^{-\Delta R}$ corrections, is to quantize the momentum (i.e., the θs) with an attendant effect upon finite-volume phase space.[21] Here however the quantization conditions are more complex than that of the TFIM model. We must take into account the nontrivial interactions between the excitations and solve the resulting Bethe ansatz equations. For the calculation at hand, we solve the one- and two-particle Bethe equations for the states $|\theta_1,a_1\rangle$ and $|\theta_3,a_3;\theta_2,a_2\rangle$. For the one-particle state, we have the free quantization condition $\theta_1=\sinh^{-1}(2\pi n/R)$ for some integer n. For the two-particle case, $\theta_{2,3}$ are quantized via $e^{iR\Delta\sinh(\theta_2)}=e^{-iR\Delta\sinh(\theta_3)}=e^{i\delta_\alpha(\theta_{23})}$, where the nontrivial phase $\delta_\alpha(\theta)$ marks the presence of interactions and depends upon the particular SU(2) representation (α=singlet/triplet/quintet) into which the two-particle state falls. Because of the presence of interactions, the finite R matrix elements of the form $\langle\theta_1,a_1|n^a(0)|\theta_3,a_3;\theta_2,a_2\rangle$ are never infinite as the θs never coincide. We again see finite R provides a clean regulation of the singularities present at $R=\infty$.

The remainder of the calculation of χ^{O_3} follows in exact analogy to the TFIM. $\chi^{O_3}(\omega,q)$ takes the form of Eq. (6) with $D^{O_3}=D^I$. In this case the expansion of $C^{O_3}(x,\tau)\equiv\langle n^z(x,\tau)n^z(0)\rangle$, $C^{O_3}(x,\tau)=\Sigma_{M,N}C^{O_3}_{M,N}(x,\tau)$ appears as

$$C^{O_3}_{M,N}(x,\tau)=\sum_{\substack{\Theta_1;\cdots;\Theta_M\\ \Theta_1';\cdots;\Theta_N'}}|\langle\Theta_1,\cdots,\Theta_M|n^z(0)|\Theta_1';\cdots;\Theta_N'\rangle|^2$$
$$\times e^{-\beta E(\theta_j)}e^{-\tau[E(\theta_i)-E(\theta_j)]+ix[P(\theta_i)-P(\theta_j)]}, \quad (10)$$

where $\Theta_n\equiv\{\theta_i,a_i\}$. The partition function here admits the expansions $Z^{O_3}=1+Z_1^{O_3}+\mathcal{O}(e^{-2\beta\Delta})$ and $Z_1^{O_3}=3\Sigma_n e^{\beta\Delta\cosh(\theta_n)}$. With these redefinitions of $C_{M,N}$, D, and Z_n, $\Sigma_1^{O_3}$ takes the same functional form as Σ_1^I.

In Fig. 2 we plot the resulting finite T DS for the O(3) NLSM. We again find that the lineshape is characterized by an asymmetry that grows with temperature. We observe that this asymmetry is far stronger than that of the semiclassical analysis (see Fig. 10 of Ref. 11) whose lineshape is well described by a Lorentzian even up to temperatures of $T=0.5\Delta$. The origin of this discrepancy between our treat-

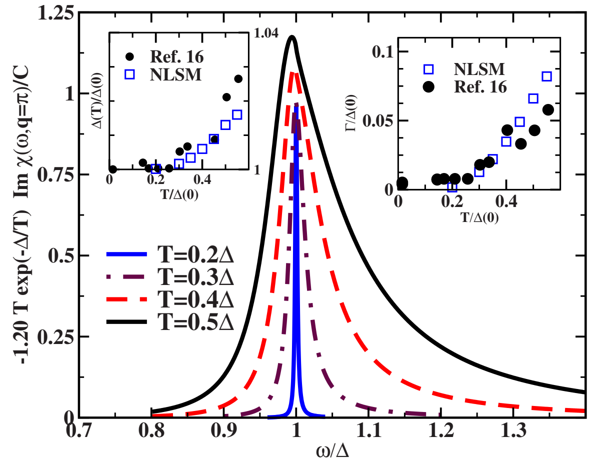

FIG. 2. (Color online) Imaginary part of $\chi(\omega,q)$ for several temperatures for the O(3) NLSM in the thermodynamic limit. Insets: temperature-dependent gap $\Delta(T)$ and width $\Gamma(T)$ (see text).

ment and the semiclassics is similar to that of the TFIM, i.e., finite-energy effects in the scattering of excitations and in the matrix elements involving the operator n_z.

The asymmetry in the lineshape can be interpreted in terms of a temperature-dependent gap $\Delta(T)$ and width $\Gamma(T)$.[12] Following Ref. 12, these are extracted as the location and half-width half-maximum of the peak of a Lorentzian fitted to the asymmetric lineshape. We plot $\Delta(T)$ and $\Gamma(T)$ vs T in the left and right insets to Fig. 2, respectively, and compare our computations with the neutron-scattering measurements performed on the spin-1 chain Y_2BaNiO_5 in Ref. 12. We see good qualitative agreement. We thus believe that our computational methodology has produced the first quantitatively accurate estimate for $\Delta(T)$. In order to facilitate further analysis of the experimental data, we suggest instead the following simple fit to the lineshape: $\mathrm{Im}\,\chi(\omega,\pi/a_0)=a/\{[\omega-\Delta(T)]^2+b\}^{1-c(\omega/\Delta-1)}$.

In conclusion we have presented a general method for the calculation of low-temperature dynamical response functions in gapped integrable models. We have applied it to calculate the lineshape of the coherent mode in gapped quantum spin chains at low temperatures. To do so, we have employed a continuum integrable representation of the chains and regulated infinities in matrix elements that appear in the infinite volume limit by working in finite but asymptotically large systems. Our method, governed by the small parameter $\exp(-\beta\Delta)$, further employs a Dyson-like resummation of the matrix elements appearing in a Lehmann expansion of the DS. As our primary conclusion, we find that the lineshape is asymmetric with an asymmetry increasing with temperature.

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