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Physics of integer-spin antiferromagnetic chains: Haldane gaps and edge states

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This article is dedicated to Michel Verdaguer, chemist, teacher, and colleague *extraordinaire*.

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ABSTRACT

The antiferromagnetic Heisenberg spin chain with *integer* spin has a short-range magnetic order and an excitation energy gap above the ground state. This so-called Haldane gap is proportional to the exchange coupling constant J of the Heisenberg chain. In this study, we discuss recent results about the spin dependence of the Haldane gap and conjecture an analytical formula valid asymptotically for large spin values. We then discuss the robustness of the edge states of the spin-1 chain by studying a spin-1 ladder using the density matrix renormalization group (DMRG) algorithm. We show that the peculiar hidden topological order of the spin-1 chain disappears smoothly by increasing the ladder rung coupling without any intervening phase transition. This is evidence for the fragile character of the topological order of the spin-1 chain.

1. The nonlinear σ model and the Haldane gap

It is well understood that the origin of exchange interactions between magnetic ions in an insulator is essentially of quantum mechanical nature. Indeed, it involves electron delocalization and the Pauli principle as needed for fermionic elementary constituents of matter. However, the description of magnetic properties of solids at the mesoscopic or macroscopic scale very often does not involve a fundamental way quantum mechanics. In case of ferromagnetic or antiferromagnetic chains, one can use coarse-grained magnetization vectors to describe ordered states that are perfectly classical quantities. Also excited states above ordered ground states like magnons admit a classical description, which has a wide range of applicability. This classical line of thought has been applied for a long time also in the realm of one-dimensional quantum spin systems with some success in the case of systems with rather large spin values. Therefore, it came as a surprise when Haldane [1–3] in the beginning of the 1980s made the bold claim that integer-spin antiferromagnetic chains have a gap to all excitations at odds with previous magnon-based reasonings. The picture that emerges is that integer-spin chains have only a short-range magnetic order even at zero temperature with spin–spin correlations decaying exponentially with a characteristic length, and there is a gap to all excitations. On the contrary, half-integer-spin chains have antiferromagnetic order with no characteristic scale albeit decaying as a power law and no gap to excited states. Interestingly, the first Haldane gap spin chain NENP [4] was synthesized roughly at the same time as Haldane's conjecture appeared. Since its synthesis, NENP has been an extremely good platform [5–7] to investigate Haldane gap physics. Quantitative comparisons between theory and experiments have been successful [8–10]. Detailed investigations have confirmed the existence of the Haldane gap close to $0.41J$, where J is the nearest-neighbor antiferromagnetic exchange of the isotropic Heisenberg chain [11–13].

The Haldane conjecture was originally based on the derivation of an effective quantum field theory using an

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expansion of fluctuation about local antiferromagnetic order of a spin-S chain. This derivation has been the subject of much theoretical work and is now available in streamlined form in textbooks [14]. In this section, we propose a quick presentation of the main arguments and proceed to discuss the dependence of the Haldane gap on the spin magnitude S, as there are now several numerical studies [15] extending up to $S = 4$.

The starting point is the imaginary time representation of the partition function in terms of spin coherent states:

$$\mathscr{Z} = \int \mathscr{D}\widehat{\Omega}_i \exp\left\{ iS\sum_i \omega[\widehat{\Omega}_i] - \int_0^\beta S^2 \sum_i \widehat{\Omega}_i \cdot \widehat{\Omega}_{+1} \right\} \quad (1)$$

The spin operators $S_i^{x,y,z}$ are written in terms of the spin coherent states $S_i^{x,y,z} \to S\widehat{\Omega}_i^{x,y,z}$, where the quantities $\widehat{\Omega}_i$ are classical commuting vectors of unit norm and S is the spin magnitude. In fact, the Haldane mapping is an asymptotic expansion valid for large S and its quantitative use for $S = 1$ is not guaranteed. Numerical direct studies down to $S = 1$ have confirmed its applicability even in this case. The first term $\sum_i \omega[\widehat{\Omega}_i]$ in the exponential is the *Berry phase* contribution, which is a nontrivial function of the imaginary time evolution of the coherent state. This term is in fact responsible for the difference between integer- and half-integer-spin chains. It is irrelevant to the bulk physics of the integer-spin chain case and it is a fortunate circumstance that provided this contribution can be dropped then the field theoretical analysis of the effective model we will derive is in fact well known. From the point of view of the effective long-distance low-energy field theory, the spin$-1/2$ spin chain is much difficult to understand even if this is the only case where exact results are known for the eigenstates by the Bethe solution. Because we are discussing only the Haldane gap, we will drop the Berry phase from now on (in the case of integer spin the Berry phase term is responsible for the appearance of edge spins $S = 1/2$, which is discussed in Section 2). We now proceed to construct an effective model valid for long distance and low energies. This requires the identification of the modes that are pertinent in this limit. We assume that these are antiferromagnetic as well as ferromagnetic fluctuations as observed in all approximation schemes used to study spin-chain physics. We write the spin coherent states as

$$\widehat{\Omega}_i = (-)^i \widehat{n}_i \sqrt{1 - \mathbf{L}_i^2 / S^2} + \frac{1}{S}\mathbf{L}_i \quad (2)$$

where $\widehat{n}_i$ is a unit vector and $\mathbf{L}_i$ is a vector perpendicular to $\widehat{n}_i$. This way of writing the spin coherent state leads naturally to a $1/S$ expansion. The nearest-neighbor Heisenberg coupling is expanded as follows:

$$\widehat{\Omega}_i \cdot \widehat{\Omega}_{i+1} = -1 + \frac{1}{2}(\widehat{n}_i - \widehat{n}_{i+1})^2 + \frac{1}{2S^2}(\mathbf{L}_i + \mathbf{L}_{i+1})^2 \quad (3)$$

The Berry phase contribution in the path integral leads to a coupling between the $\mathbf{L}$ vector and the imaginary time derivative of the order parameter field $\widehat{n}_i$:

$$\int \mathscr{D}\mathbf{L}_i^\perp \exp\left\{ -\int_0^\infty d\tau \int \frac{dq}{2\pi} (\widehat{n} \times \partial_\tau \widehat{n})_{-q} \cdot \mathbf{L}_q - \int_0^\infty d\tau \int \frac{dq}{2\pi}\, \chi_q^{-1}\, \mathbf{L}_q \cdot \mathbf{L}_{-q} \right\} \quad (4)$$

where the susceptibility is given by $\chi_q^{-1} = 4J\cos^2(qa/2)$, with a being the distance between spins. Progress can be made if we take the long wavelength limit $q \to 0$ in the susceptibility $\chi^{-1} \to 4J \equiv \chi_0$. Then, we can integrate the ferromagnetic fluctuation vector $\mathbf{L}$ and obtain a simple quadratic action:

$$\mathscr{Z} = \int \mathscr{D}\widehat{n} \exp\left\{ -\frac{1}{2}\int_0^\infty d\tau \int dx \left[\chi_0 (\partial_\tau \widehat{n})^2 + \rho_s (\partial_x \widehat{n})^2 \right] \right\} \quad (5)$$

where we have introduced a stiffness $\rho_s = JS^2 a$. Now Euclidean time and space are on an equal footing, provided we rescale by the velocity $c = \sqrt{\rho_s/\chi_0}$. At zero temperature, $\beta \to \infty$; this field theory is in fact the well-known nonlinear σ model (NLσM). It was introduced to describe critical properties of classical Heisenberg ferromagnets at nonzero temperatures. Here, we observe the consequence of the mapping of a $1+1$ dimensional quantum system onto a classical two-dimensional system. In this new language, there is a very basic fact that there is no order at any nonzero temperature for classical Heisenberg spins. In physical terms, spinwaves disorder the system beyond a finite correlation length ξ and spin correlations decay exponentially with distance. We thus infer immediately that the space correlations of the $\widehat{n}$ vector field decay exponentially. In addition, as we have complete equivalence between space and (Euclidean) time, the time correlations also decay exponentially:

$$\langle \widehat{n}(0,x)\widehat{n}(\tau,x)\rangle \approx \exp\left(-(c\tau)\xi^{-1}\right) \quad (6)$$

It is a classic result in Euclidean quantum mechanics that the imaginary-time decay of correlations is governed by the energy gap of the system. Thus, we conclude from Eq. 6 that the spin chain has a gap $\Delta = c\xi^{-1}$. If we rewrite the effective action in terms of the conventional NLσM, we find the action as follows:

$$\mathscr{S}_{\mathrm{NL\sigma M}} = \frac{1}{2g}\int d\tau\, dx\, (\partial_\mu \widehat{n})^2 \quad (7)$$

with the coupling constant $g = c/\rho_s = 2/S$. Renormalization group calculations have shown that the correlation length of this model has a known dependence upon g:

$$\xi = C\frac{2\pi}{g}\exp(-2\pi/g)(1 + O(g)) \quad (8)$$

where C is a pure number. Because we know the spin dependence of the velocity $c = 2JSa$, we deduce that the gap depends upon the spin value as

$$\Delta_S / J = C_0 S^2 \exp(-\pi S)(1 + O(1/S)) \quad (9)$$

with a prefactorC_0, which is a pure number independent of the spin. This formula should be understood as being asymptotic, that is, valid when $S \to \infty$. The case $S = 2$ has been studied experimentally [16] and theoretically [17]. More recent theoretical estimates are available for $S = 3$ [18] and even for $S = 4.10$ cm [15].

The pure number in factor of the σ model formula for ξ is known and is $C = e/8$ for a specific choice of the renormalization procedure. The renormalization procedure corresponding to the quantum lattice model cannot be easily related to the standard ways of renormalizing the σ model. Because we are not yet able to make this correspondence precise, we choose to make an extrapolation from known values of the Haldane gap up to $S = 4$ (see Table 1). The Shanks extrapolation for three numbers a, b, c is simply calculated by the ratio $(ac - b^2)/(a + c - 2b)$ and is given in the last column of Table 1. After some trial and error, we are led to propose that the limiting value is $2\exp(2) = 14.778\ldots$ close to 14.525. We thus conclude this section by conjecturing the asymptotic formula for the Haldane gap

$$\Delta_S / J = 2S^2 \exp(2 - \pi S)(1 + O(1/S)) \tag{10}$$

It remains to be confirmed whether methods like those of ref. [19] can relate the renormalization schemes and confirm this prefactor.

2. Edge states of a Haldane ladder

Another important feature of the spin-1 chain is the existence of a hidden magnetic order, which is long range. We have already commented on the fact that ordinary spin correlations decay exponentially with a finite correlation length. However, there is nevertheless a hidden order in the system. The nearest-neighbor Heisenberg $S = 1$ spin chain is not a solvable system, and hence one has to use numerical methods to study its quantitative properties. However, it is possible to perturb the Hamiltonian and obtain a new model [20] called AKLT whose ground state wavefunction can be found by elementary means, although it is still not solvable. The trick is to add a biquadratic spin coupling:

$$\mathscr{H}_{\mathrm{AKLT}} = \sum_i S_i \cdot S_{i+1} + \frac{1}{3}(S_i \cdot S_{i+1})^2. \tag{11}$$

Table 1
Gaps of the spin-S Heisenberg chain using state-of-the-art numerical results [15].

S	Gap/J	NLσM	Gap/NLσM	Shanks
1	4.104800×10^{-1}	4.321392×10^{-2}	9.499	–
2	8.916000×10^{-2}	7.469771×10^{-3}	11.936	–
3	1.002000×10^{-2}	7.262957×10^{-4}	13.796	19.787
4	7.990000×10^{-4}	5.579748×10^{-5}	14.320	14.525

The first column gives the spin value, the second column the most accurate known values of the dimensionless gap, and the third column the bare σ model number from which we compute ratio in the fourth column. It should extrapolate to a constant for infinite spin. The extrapolation is performed in the last column using the simple three-term Shanks extrapolation.

The motivation is formal, because in the real world, a superexchange theory leads to negligible higher-spin couplings. However, with the very special choice of the 1/3 coefficient for the biquadratic term, the spin operator is now the projector onto the total spin $S = 2$ of the pair $i, i+1$. This can be checked by elementary means (a bit tedious). Now we proceed to exhibit a wavefunction, which is an exact zero-energy eigenstate of Hamiltonian (Eq. 11). Each local spin-1 can be viewed as the triplet state of two spin-1/2 residing at the same site i. In the case of NENP, this corresponds to microscopic physics, because spin-1/2 elementary electrons in the nickel ions are coupled by Hund's coupling with a triplet state. Now, we make singlet bonds between neighboring sites (ions) and construct the wavefunction by capturing all spins in the following way:

$$|\boldsymbol{\Psi}_{\mathrm{AKLT}}\rangle = \ldots \otimes \{|\uparrow_i \downarrow_{i+1}\rangle - |\downarrow_i \uparrow_{i+1}\rangle\} \otimes \{ \\ \times |\uparrow_{i+1} \downarrow_{i+2}\rangle - |\downarrow_{i+1} \uparrow_{i+2}\rangle\} \otimes \ldots \tag{12}$$

If we consider two neighboring spins, then the total spin of such a pair can only be 0 or 1 but not 2, because the central spins 1/2 are already engaged in a singlet state. Thus, we see that all projectors onto the $S = 2$ state for any pair gives simply zero and hence, this wavefunction is an exact zero-energy eigenstate of the Hamiltonian (Eq. 11). In fact, this is the exact ground state of the AKLT Hamiltonian. Although this construction may seem artificial, we note that this Hamiltonian can be viewed as a perturbation of the usual Heisenberg Hamiltonian provided “1/3 is small” meaning that the biquadratic operator does not change the physics of the system. Indeed, we are lucky and it has been shown in detail that this is the case [21]. As a consequence, reasonings based on the model AKLT state are likely to be correct for the real world dominated by the Heisenberg exchange. A very simple inference from the AKLT description is that if we consider an open chain, then there are dangling spins 1/2 at the end. Of course real materials always involve open chains and we thus can predict that local probes should detect effective spin 1/2 degrees of freedom at the end of NENP chains. The experiment has been done [22] using NMR of a zinc impurity breaking the chain and the number of NMR lines is exactly in agreement with the prediction of the spin 1/2 instead of 1 as one may guess naively. For an open chain, we expect that the two end spins 1/2 appear as almost degenerate singlet and triplet states due to a coupling through the bulk of the chain that should go exponentially to zero when the chain length increases. So the spectrum of an open chain should display quasidegenerate $S = 0$ and $S = 1$ states and, above a (Haldane) gap, an $S = 2$ excitation will be obtained by creating a magnon-like state [23]. If we look at the magnetization profile of the $S^z = 1$ member of the triplet, it should display nonzero magnetization only close to the edges of the chain. This is indeed what is observed numerically for the $S = 1$ chain using the density matrix renormalization group (DMRG) algorithm [13,24].

We now turn to the question of the robustness of the formation of these end spin states. They are due to the special ordering pattern described by the AKLT wavefunction [20,25], and it is still present for the realistic

Heisenberg exchange Hamiltonian. It is known that several physical effects can destroy the Haldane gap, such as too strong anisotropies either local on-site or exchange, and also an applied magnetic field can close the Haldane gap. If we cross a phase transition by such a mechanism, we do not expect edge states to survive because the peculiar long-range correlations of the AKLT state are destroyed. It has been noted some time ago that in fact there is no need for a sharp phase transition to kill the hidden order and the edge states. This is very clear in the case of the magnetization process of the Heisenberg $S = 1$ chain. Indeed, under an applied field the Haldane gap goes to zero at some critical field H_c and the magnetization remains zero up to this value. When the critical value is exceeded, the magnetization starts to increase continuously up to full saturation and the edge states disappear right at H_c. If now we consider a realistic material like NENP, then there are small but nonzero anisotropies that break the spin rotation symmetry. As a consequence, the magnetization transition [26] is now rounded and is no longer sharp. Magnetization appears immediately for infinitesimal applied fields and edge states disappear when going to saturation without any phase transition. This phenomenon has been dubbed [27] “symmetry protected topological order”. It leaves open the possibility that the hidden order and accompanying edge states may disappear even without breaking any symmetry and without any phase transition. We now show that this is the case by studying a coupled $S = 1$ ladder system. We thus envision a plausible molecular magnet in which neighboring Haldane chains of spins $S = 1$ are coupled along the rungs of a ladder by some independent exchange $J_\perp$. A spin Hamiltonian can thus be written as follows:

$$\mathscr{H}_{\text{ladder}} = \sum_i S_{i,A}S_{i+1,A} + \sum_i S_{i,B}S_{i+1,B} + J_\perp \sum_i S_{i,A}S_{i,B} \tag{13}$$

where the sites along the two chains are labeled by i and A, B label the two coupled chains. We have taken the exchange along the chain as the unit of energies so the only remaining parameter is the ratio of exchange interactions along and across the chains called $J_\perp$. We note that recent experiments have shown that the organic molecular magnet BIP-TENO is precisely a spin-1 ladder [28,29]. However, the exchange interactions in BIP-TENO are more complex than our simple Hamiltonian (Eq. 13).

If we consider a ladder with periodic boundary conditions, then it has been established that there is no phase transition [30,31] between the $J_\perp = 0$ decoupled chain limit and the $J_\perp = \infty$ with extremely strong rungs. This strong rung limit is simple to analyze: the ground state is made of singlet states involving two spins that are related by the rung coupling and the total state is just the tensor product of such singlets. The first excited state is made by breaking a rung bond from singlet to triplet, and this triplet will have a dispersion along the chain whose magnitude is given by the (relatively) small coupling along the chains. The small $J_\perp$ limit is two weakly coupled Haldane chains and because they are gapped, they will be resilient to any small local perturbation like a small rung coupling. Even if there is no transition between these two limits, it remains unclear what happens to the edge states of an open ladder.

We first enumerate the low-lying states of this spin system starting from our knowledge of the $S = 1$ chain. We know that each chain will have a singlet and a triplet of low-lying states separated by the Haldane gap from the higher excitations [23]. With two chains, this means a total of 16 states that can be classified as two $S = 0$ singlets, three $S = 1$ triplets, and one $S = 2$ quintuplet. When adding a very small $J_\perp$, these will no longer be degenerated and first-order perturbation theory will lead to an order $O(J_\perp)$ splitting of the states. Their edge nature can be revealed by computing the magnetization profile $\langle S_n^z \rangle$ along one of the coupled chains. We have performed DMRG calculations of this system to obtain the ground state wavefunction. Convergence becomes problematic when $J_\perp$ is very small $J_\perp < 0.001$, but results are reliable for higher values of the coupling. We use up to 1500 states per block and chain length up to 200 spins.

If we consider the low-lying $S^z = 0$ (where S^z is the total spin) state, we observe nothing remarkable. The magnetization is uniformly vanishing and this does not change from the decoupled limit $J_\perp = 0$ till $J_\perp$ reaches a very large number as expected. We now compute the ground state wavefunction in each sector $S^z = 1, 2, 3$; we expect that $S^z = 1, 2$ display only edge modes whereas $S^z = 3$ should capture a bulk magnon state. We vary the rung coupling in the $S^z = 1$ sector in Figs. 1–3. For small $J_\perp$, we observe very clear edge mode oscillations of the local magnetization and when the chain is long enough, the bulk magnetization is zero (green line in the figure), see Fig. 1. If we increase $J_\perp$, there is coexistence of edge oscillations and bulk magnetization (see Fig. 2). Finally, at large enough $J_\perp$ values, all edge phenomena are washed out and we observe a single bump of the magnetization. We interpret this state as a single magnon state of the infinite rung limit that propagates as a particle in a box as reported in a study by White and Huse [13]. The

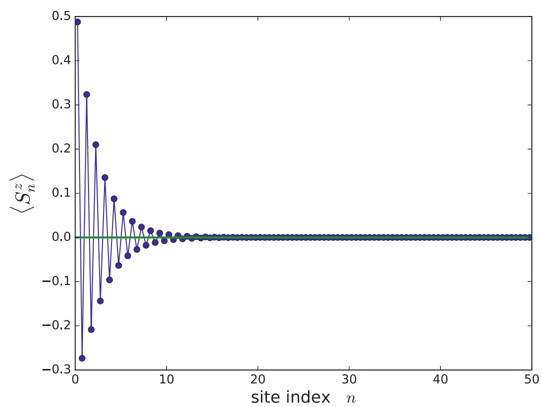

Fig. 1. The magnetization profile $\langle S_n^z \rangle$ for site n along the first 50 spins of an $S = 1$ Heisenberg ladder of 100 sites long (for a total of 200 spins). The expectation value is computed in the lowest-lying $S^z = 1$ state, which belongs to the manifold of edge states. The rung exchange coupling is $J_\perp = 0.003$. The green line is zero magnetization. The bulk is not magnetized, proving the edge nature of the excitations.

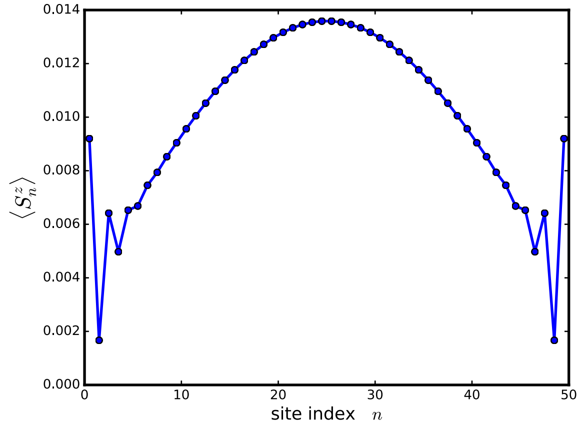

Fig. 2. The magnetization profile $\langle S_n^z \rangle$ in the lowest-lying $S^z = 1$ state for site n along the $S = 1$ Heisenberg ladder of 50 sites long (total 100). The rung exchange coupling is now $J_\perp = 0.5$; we observe that the magnetization is now essentially in the bulk and there are still edge oscillations of the magnetization.

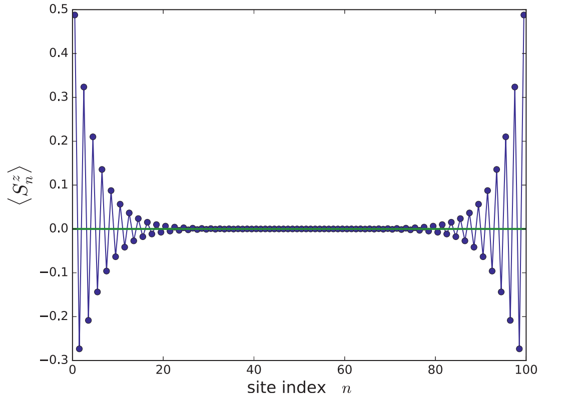

Fig. 4. The magnetization profile $\langle S_n^z \rangle$ for site n along the $S = 1$ Heisenberg ladder of 100 sites long. The expectation value is computed in the lowest-lying $S^z = 2$ state, which belongs to the manifold of edge states. The rung exchange coupling is $J_\perp = 0.003$; there is zero bulk magnetization and only edge modes.

important information is the gradual disappearance of the edge modes without any level crossing.

We expect that the same scheme applies for the lowest-lying $S^z = 2$ state. Its behavior is displayed in Figs. 4–7. We again observe clear edge modes exponentially localized at the end of the ladder with no bulk magnetization at small $J_\perp$. There is a smooth crossover to a regime that we interpret as a two-magnon state (see Fig. 7).

The situation should be different for the $S^z = 3$ sector because in the decoupled limit $J_\perp = 0$, one is forced to excite at least one magnon in one of the chains. This magnon will be delocalized and its total magnetization should spread all over the chain. This is exactly what we observe in Figs. 8 and 9. The bulk is always magnetized even when $J_\perp \to 0$. In addition to this magnon contribution, there are also edge oscillations. When increasing the coupling, the magnon contribution becomes clearer (see Fig. 9). Finally, for very large rung coupling, Fig. 10, we are left with only rung magnon excitations and no edge

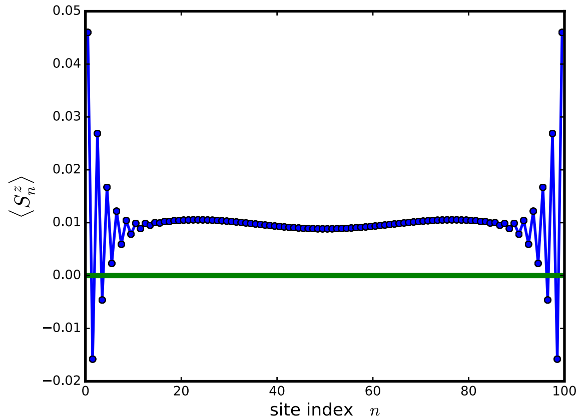

Fig. 5. The magnetization profile $\langle S_n^z \rangle$ as in Fig. 4. The rung exchange coupling is $J_\perp = 0.015$; there is now coexistence of a small bulk magnetization and edge modes.

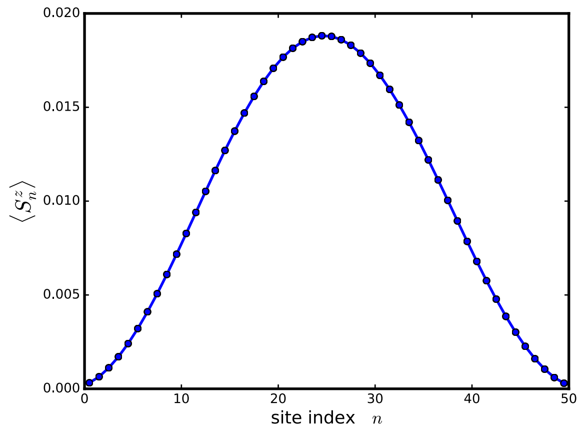

Fig. 3. The magnetization profile as in Fig. 2. The rung exchange coupling is $J_\perp = 4$; the state can be interpreted as a single magnon state clearly defined from the infinite rung limit. There is no reminder of the edge modes.

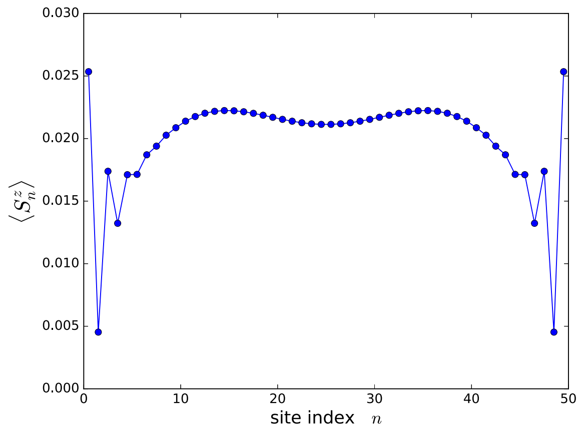

Fig. 6. The magnetization profile $\langle S_n^z \rangle$ as in Fig. 5. The rung exchange coupling is $J_\perp = 0.5$; the bulk excitations display a two-rounded-peak structure.

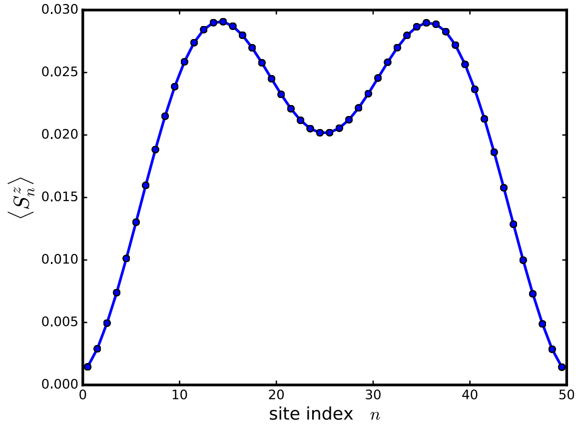

Fig. 7. The magnetization profile $\langle S_n^z \rangle$ as in Fig. 6. The rung exchange coupling is $J_\perp = 4$; edge modes have disappeared completely and we observe two magnons with characteristics of a particle-in-a-box wavefunction.

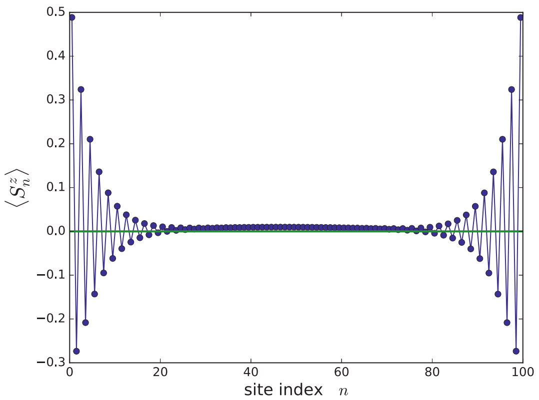

Fig. 8. The magnetization profile $\langle S_n^z \rangle$ in the lowest-lying $S^z = 3$ state. The rung exchange coupling is $J_\perp = 0.003$; although there are edge oscillations, we note that the bulk magnetization is nonzero. This is not a pure edge state.

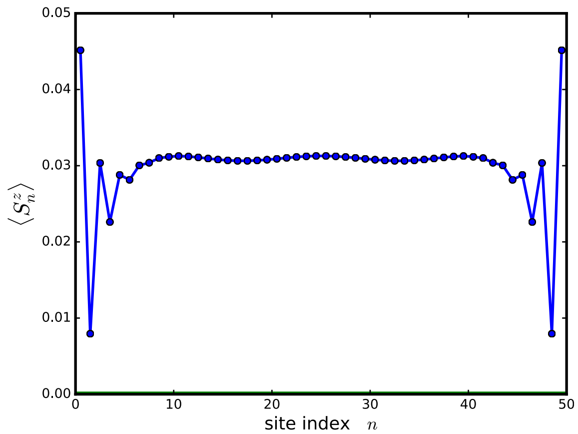

Fig. 9. The magnetization profile $\langle S_n^z \rangle$ in the lowest-lying $S^z = 3$ state. The rung exchange coupling is $J_\perp = 0.5$; edge oscillations are still present but the three-peak structure appears.

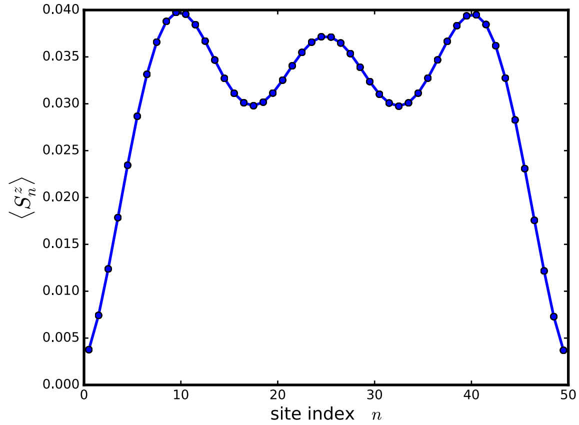

Fig. 10. The magnetization profile $\langle S_n^z \rangle$ in the lowest-lying $S^z = 3$ state. The rung exchange coupling is $J_\perp = 4$; we observe a pure three-magnon state.

modes. This is a gradual phenomenon with no physical discontinuity.

In principle, NMR on well-chosen chain-breaking impurities should be able to measure these physical effects.

3. Conclusion

Antiferromagnetic spin chains are a fascinating playground for states of magnetic matter that do not fit into the usual symmetry-breaking views of magnetism. The crafting of molecular magnets has allowed chemistry and physics to join forces and investigate experimentally the Haldane conjecture that predicts a fundamental difference between integer- and half-integer-spin chains. In the case of integer-spin chains, the so-called Haldane weakens with the spin value, and we have proposed here a conjecture for its asymptotic behavior for large spin S. Another very special feature of the Haldane gap state is the existence of long-range correlations that lead to edge modes, which have been observed in experiments [22]. These correlations are fragile and can be destroyed even without crossing any phase transition. We have given an explicit mechanism for the destruction of the hidden order by showing that an $S = 1$ spin ladder interpolates between a haldane-style phase and a simple tensor product phase with no kind of long-range order. The crossover we describe should be accessible to NMR measurements, provided one finds the appropriate molecular magnet. In this respect, the recent studies of BIP-TENO [28,29] or other magnets [32] show that it is indeed feasible to study spin-1 ladders and observe the interesting crossover between the Haldane phase and the rung singlet phase.

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