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Antiferromagnetic Heisenberg spin-1 chain: Magnetic susceptibility of the Haldane chain described using scaling

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The phenomenological expression $\chi T/(Ng^2\mu_B^2/k)=C_{1n}\exp(-W_{1n}/T)+C_{2n}\exp(-W_{2n}/T)$ describes very accurately the temperature dependence of the magnetic susceptibility computed for antiferromagnetic rings of Heisenberg spins $S=1$, whose size n is even and ranges from 6 to 20. This expression has been obtained through a strategy justified by scaling considerations together with finite size numerical calculations. For n large, the coefficients of the expression converge towards $C_1=0.125$, $W_1=0.451J$, $C_2=0.564$, $W_2=1.793J$ (J is the exchange constant), which are appropriate for describing the susceptibility of the spin-1 Haldane chain. The Curie constant, the paramagnetic Curie–Weiss temperature, the correlation length at $T=0$ and the Haldane gap are found to be closely related to these coefficients. With this expression, a very good description of the magnetic behavior of Y_2BaNiO_5 and of $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP), the archetype of the Haldane gap systems, is achieved over the whole temperature range.

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I. INTRODUCTION

The antiferromagnetic (AF) chain of Heisenberg spins $S=1$ is still an object of interest many years after Haldane conjectured that an energy gap Δ separates the singlet ground state from the first excited state[1] for all AF chains with integer spin values, while the excitation spectra of the half integer spin chains are gapless. The Haldane prediction was confirmed experimentally and theoretically.[2–4] After the pioneering work of Botet *et al.*, who solved numerically the spin Hamiltonian for finite rings of n spins with n up to 12, further calculations have been performed by means of the quantum Monte Carlo[5] and the density-matrix renormalization group (DMRG) techniques[6] to determine more precisely the energy gap as n diverges. The extrapolated Haldane gap value for the infinite chain was estimated to be $\Delta\sim 0.41J$,[4] where J is the nearest neighbor exchange interaction.

The spin-1 system displays other fascinating features as well. For example, the ground state of an open spin-1 open chain is characterized by an effective spin $S=1/2$ at each end, which leads to fourfold degeneracy of the ground state. Furthermore, the correlation length at $T=0$ reaches a finite value, $\xi=2JS/\Delta$, which directly results from effects of spin fluctuations in low dimension.

In this paper, we derive a phenomenological expression of the magnetic susceptibility (χ) of AF rings made up of an even number n of Heisenberg spins $S=1$ which, extrapolated to the thermodynamic limit, captures the main features of the Haldane chain. The method is based upon physical scaling arguments[7] that we recall in Sec. II and it is applied to the theoretical data on finite quantum rings in Sec. III. A description of the numerical methods used for solving the problem of the quantum Heisenberg rings is given in the same section. The whole procedure provides a new insight upon the gap and the finite correlation length at $T=0$, and ultimately it results into a formula which is also useful to fit the experimental evidence, as demonstrated for two Haldane chain compounds, Y_2BaNiO_5 (Ref. 8) and $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP). A brief physical interpretation of the results is attempted in Sec. IV.

II. NON-CRITICAL-SCALING: THE OTHER SOLUTIONS OF THE SCALING MODEL

Let $\chi T=Np_{\mathrm{eff}}^2/3k$, where $p_{\mathrm{eff}}=g\mu_B[S(S+1)]^{1/2}$, be the Curie constant of N independent atoms of size ξ_0. In the presence of interactions between the atoms, larger units, whose size $\xi(T)$ defines the correlation length, act as the new independent entities. By definition, the number of these units is $N(T)\propto\xi(T)^{-D}$ in space dimension D. We assume, furthermore, following Néel in his approach of the superparamagnetism of fine AF particles, that the effective moment of each unit increases like a power D' which may differ from the space dimension D.[7] Then $p_{\mathrm{eff}}\propto\xi(T)^{D'}$ and

$$\chi T\propto\xi(T)^{-(D-2D')}. \tag{1}$$

We now assume that $\xi(T)$ increases when T decreases, and that it is possible, in the spirit of Kadanov's renormalization scheme, to connect by a "hierarchical recipe" the successive steps of the cascade which relates $\xi(T)$ and ξ_0. Assuming, for example, that ξ is multiplied by b when $(J/T_C-J/T)$ is multiplied by a, then, if n steps are needed to relate T_n with J, we can write[7]:

$$J/T_C - J/T_n = a \times (J/T_C - J/T_{n-1}) = a \times a \times a \cdots \times (J/T_C - 1) = a^n \times (J/T_C - 1), \quad (2)$$

$$\xi_n = b \times \xi_{n-1} = b \times b \times b \times \cdots \times \xi_0 = b^n \times \xi_0. \quad (3)$$

By eliminating n between Eqs. (2) and (3), one derives the standard relation of static scaling:

$$\xi(T)/\xi_0' = (1 - T_C/T)^{-\nu} = (1 - T_C/T)^{-\Theta/T_C}. \quad (4)$$

We have introduced above $\xi_0' = (1 - T_C/J)^{-\nu}$, $\nu = -\log(a)/\log(b)$ and (or) $\Theta = \nu T_C$, which must be positive in order to ensure that $\xi(T)/\xi_0' = 1 + \Theta/T + \cdots$ is an increasing function of $1/T$. The "static scaling assumption" has measurable consequences. In particular, using Eq. (1) and allowing T_C to be positive null or negative, we find[7]:

$$\chi T/(Ng^2\mu_B^2/k) = C \times (1 - T_C/T)^{-(2D'-D)\nu} = C \times (1 - T_C/T)^{-\gamma} \text{ for } T_C > 0, \quad (5)$$

$$\chi T/(Ng^2\mu_B^2/k) = C \times \exp((2D' - D) \times \Theta/T) = C \times \exp(-W/T) \text{ for } T_C = 0, \quad (6)$$

$$\chi T/(Ng^2\mu_B^2/k) = C \times (1 + T_K/T)^{(2D'-D)\Theta/T_K} = C \times (1 + T_K/T)^{-\gamma} \text{ for } T_C = -T_K. \quad (7)$$

The solutions (5) are the familiar power laws appropriate to describe the usual phase transitions, with $\gamma = (2D' - D)\nu$ being the critical exponent related to the spin and lattice dimensionalities. Note that instead of T/J, we have used J/T (which goes to zero as T diverges) to construct the scaling variable $(J/T_C - J/T)$. This is to ensure that all three equations above have a sensible high temperature expansion, where the Curie–Weiss law, $\chi = C/(T-W)$, is recovered, with $C = S(S+1)/3$ being the Curie constant, and $W = (2D' - D)\Theta$ the paramagnetic Curie temperature in all three cases above.

Let us now focus upon other aspects of the above equations that have, in general, been left aside. First, we observe that the solutions of type (6) or (7) have the same legitimacy as the solutions of type (5) and that they are not forbidden by any thermodynamic rule. They are therefore natural candidates to describe those systems, sitting at or below a "lower critical dimensionality," where spin correlations are significant but where no long-range order sets in at any finite temperature. Second, we observe that, for a given positive $\Theta = \nu T_C$, the model provides, depending on the sign of $2D' - D$, both ferromagnetic solutions where χT increases upon cooling down and AF solutions where χT decreases. We intend to use the latter to describe the magnetization of real AF systems, in rupture with a tradition whereby it is believed that the usual susceptibility does not contain valuable information, and that the "staggered" magnetization should be considered instead, and discussed in the same manner as the magnetization in a ferromagnetic system.

In order to check these ideas and to decide which expression is more appropriate to describe experimental systems, we propose to differentiate Eqs. (5)–(7) to obtain the equivalent expression:

$$\partial \log(T)/\partial \log(\chi T) = -(T - T_C)/\gamma T_C = -(T - T_C)/W. \quad (8)$$

It appears that $\partial \log(T)/\partial \log(\chi T)$ is a linear function of T with slope $-W^{-1}$, in the temperature window where the scaling argument is valid, and that γ^{-1} and T_C are simultaneously deduced from the intersection of the straight line with the axes. In the $T_C = 0$ limit, where the χT product is given by Eq. (6), the straight line intersects the axes at their origin.

In previous works, we have reported examples where $\partial \log(T)/\partial \log(\chi T)$ obtained by differentiating the experimental data shows a unique linear regime, giving a T_C value that is positive null or negative.[7,9,10] In some cases, we observe an abrupt crossover, from one regime to the other [e.g., from solutions of type (6) to solutions of type (5)] occurring at a particular temperature where the effective space dimensionality of the system changes (e.g., because correlations from chain to chain which were negligible at the scale of atomic distances in a linear system may become important on larger segments).[10]

We will, hereafter, consider, still, another situation where two different criticalities seem to coexist in the same temperature range, i.e., where two solutions of type (6) are superimposed in a common temperature window.

III. APPLICATION OF THE MODEL TO THE SPIN-1 HALDANE CHAIN

Since the work of Botet *et al.* on the ground-state properties of finite AF rings with $n = 6$ to 12 spin-1 sites, using Lanczös' algorithm,[2] many numerical calculations have complemented our knowledge of the physics of the Haldane chain, focusing in particular on the spectrum of low-lying spin states, with a special emphasis on the energy gap, the exponential decay of the spin correlation function and the correlation length as $T \to 0$.[4]

We have, hereafter, obtained the thermodynamic properties of the finite AF rings of n spins $S = 1$ by the exact diagonalization of the spin Hamiltonian for n up to 10 and by the density-matrix renormalization group (DMRG) technique for $n = 16$ and 20. Periodic boundary conditions have been imposed to minimize finite size effects, while only even n values have been considered to avoid frustration effects.

A standard DMRG procedure was used to construct the spin Hamiltonian matrices in the DMRG basis, imposing periodic boundary conditions to minimize the finite size effects with $S_{N+1} = S_1$.[6,11] It might appear surprising that the DMRG method, in which one or a few states only, usually the ground state and one or two excited states, are targeted in each iteration, can provide accurate thermodynamic properties. It is, however, well known that, at each iteration, the DMRG space, which contains the lowest energy state, has substantial projections from low-lying excited states. Hence, these excited states can be well described in the chosen DMRG basis. Furthermore, the low-energy states of the full system, which are important from the low-temperature thermodynamic

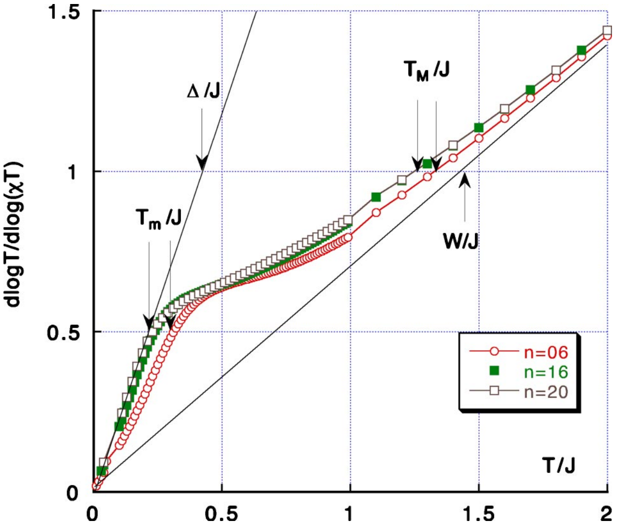

FIG. 1. (Color online) $\partial \log(T)/\partial \log(\chi T)$ deduced from exact calculations of the susceptibility of AF Heisenberg rings of n spins $S=1$ is shown vs T/J for $n=6$, 16, and 20. The linear regime at high temperature, which extrapolates to the origin of the axes, points an exponential solution of type (6) for T larger than T_M, where $\partial \log(T)/\partial \log(\chi T)=1$. A distinct exponential regime is observed at temperatures smaller than T_m where $\partial \log(T)/\partial \log(\chi T)=0.5$. The low temperature gap saturates to the Haldane gap value Δ when n increases.

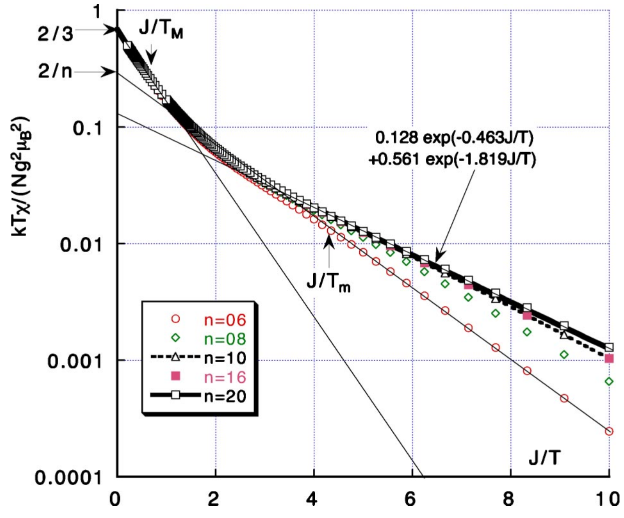

FIG. 2. (Color online) Results of exact calculations of the susceptibility $\chi(T)$ of AF rings of n Heisenberg spins $S=1$, for n equal to 6, 8, 10, 16, and 20. For $T>T_M$, there is a high temperature Arrhenius regime, whereby the χT data first depart from the theoretical Curie constant for $S=1$ $\left(C_H=\frac{2}{3}\right)$. Below T_m, there is a distinct low temperature Arrhenius regime whose Curie constant is $C_{Ln}=2/n$ and whose activation energy W_{Ln} is the Haldane gap. The different parameters C and W for all n are listed in Table I.

point of view, are often found to be few of the low-energy states in different total S^z sectors. We first set up the Hamiltonian matrices for all the allowed total S^z sectors for a ring of 20 spin-1 sites. We then diagonalize these matrices to obtain all the eigenvalues in each of the total S^z sectors. The number of direct product states increases roughly as $9m^2$, for a density-matrix cut-off of m in a spin-1 chain. As the number of DMRG basis states increases rapidly with increasing m, and since we have to diagonalize the matrices fully, we retain a smaller number of dominant density matrix eigenvectors in the DMRG procedure, i.e., $60 \leqslant m \leqslant 80$, depending on the total S^z sectors. These energies, however, turn out to be quite accurate, even on an absolute scale.[12] Calculations of thermodynamic properties from these eigenvalues are then quite straightforward.

Figure 1 shows $\partial \log(T)/\partial \log(\chi T)$ vs T, which are obtained by differentiating numerically the $\log(\chi T)$ vs $\log(T)$ data computed for AF rings of n spin-1 sites, with $n=6$ to 20. In both the high and the low temperature regimes, the data are positive and approach a straight line which intersects the axes near the origin at $T=0$. This indicates that an AF exponential solution of the type (6) is relevant in either range. The high temperature solution $\chi T = C_{Hn} \exp(-W_{Hn}/T)$ is much the same for all n values, and valid at temperatures larger than T_M where $\partial \log(T)/\partial \log(\chi T)=1$ (this is where the magnetic susceptibility χ is maximum). The low temperature solution $\chi T = C_{Ln} \exp(-W_{Ln}/T)$ is valid at temperatures smaller than T_m where $\partial \log(T)/\partial \log(\chi T)=0.5$ (this is where χ/T is maximum). The high and low temperature regimes are described by different straight lines for $T>T_M$ and $T<T_m$ in the Arrhenius plot of Fig. 2. The same data are shown in Fig. 3 as $\chi(T)$ vs $\log T/J$. The corresponding parameters C and W are listed in Table I and their dependence on $1/n$ is displayed in Fig. 4.

For $n \leqslant 10$, each parameter is precisely what the theory predicts. The Curie constant in the high temperature regime agrees with the expected $S(S+1)/3$ value. The associated activation energy, namely, the actual paramagnetic Curie–Weiss temperature, approaches $W_H=1.44J$ for all n, which is close enough to the high temperature limit $4J/3$ deduced analytically for a ring of 4 spins. In the low temperature regime, we have $C_{Ln}=2/n$, which is the expected value for a

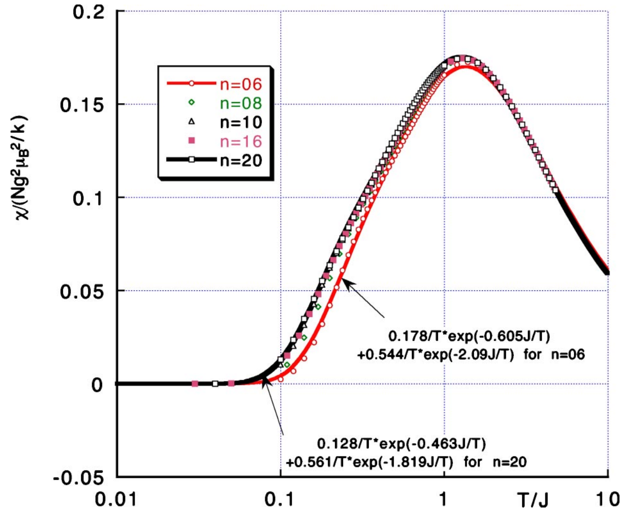

FIG. 3. (Color online) The same susceptibility data as in Fig. 2 are shown here versus $\log(T)$, with the best values of the coefficients C_{1n},W_{1n} and C_{2n},W_{2n}, characterizing the low and the high temperature regimes in a single fit.

TABLE I. Exact value of the gap $\Delta(n)$ between the ground and the first excited state, and best values of the parameters of Eq. (9) for AF rings of n Heisenberg spins $S=1$. The best values of the same parameters characterizing the high and the low temperature regimes in a single fit (see Fig. 3) are also listed.

n	$1/n$	$\Delta(n)/J$	C_{Ln}	W_{Ln}/J	C_{Hn}	W_{Hn}/J	C_{1n}	W_{1n}/J	C_{2n}	W_{2n}/J
4	0.2500	1.000	0.486	0.999	0.680	1.495	0.204	0.769	0.506	2.087
6	0.1666	0.721	0.329	0.720	0.674	1.444	0.178	0.605	0.544	2.090
8	0.1250	0.594	0.248	0.593	0.673	1.438	0.145	0.512	0.558	1.916
10	0.1000	0.525	0.193	0.524	0.673	1.439	0.132	0.475	0.561	1.844
12	0.0833	0.502								
14	0.0714	0.486								
16	0.0625	0.478	0.162	0.483	0.673	1.437	0.132	0.475	0.561	1.844
18	0.0555	0.469								
20	0.0500	0.457	0.191	0.523	0.673	1.438	0.128	0.463	0.561	1.819
∞	0.0000	0.421					0.125	0.451	0.564	1.793

singlet-triplet spin configuration well separated from the high spin high-energy states (in that temperature range, the influence of upper excited states is negligible). The associated W_{Ln} is precisely the energy distance $\Delta(n)$ from the fundamental to the first excited level, that is directly deduced by computation for finite chains and is also reported in Table I. $\Delta(n)$ decreases as n increases and its dependence on n is well approximated by the power law $\Delta(n)=0.421+6.5n^{-1.74}$. In the $n\rightarrow\infty$ limit, it tends towards a finite value that may be compared to the Haldane gap. Note that our finite size calculations overestimate the extrapolated χ at $T=0$ and so the gap; however, the finite size numerical study has been carried out in the spirit of comparison with the scaling analysis.

This remarkable aptitude of two distinct exponentials for representing the magnetic susceptibility in two different temperature ranges was a motivation to fit the data at all temperatures from two superimposed exponentials. The optimization has been achieved with $\chi(T)$ rather than the $\chi T(T)$, because more weight is given to the data in the range of the susceptibility maximum. Clearly the C coefficients given in Table I need be related by $C_{Hn}\approx C_{1n}+C_{2n}$ and $C_{Hn}W_{Hn}\approx C_{1n}W_{1n}+C_{2n}W_{2n}$ to reproduce the main features in the high temperature limit. Similarly, $C_{Ln}\approx C_{1n}$ and $W_{Ln}\approx W_{1n}$ in the low temperature regime where one exponential necessarily dominates. These constraints are pretty well realized with the C's and W's given in Table I. The resulting fit is very good (Figs. 2 and 3), and the parameters become nearly constant for rings larger than 10 spins, as shown in Fig. 5. For such rings, we propose the following expression of the χT product (normalized to $Ng^2\mu_B^2/k$),

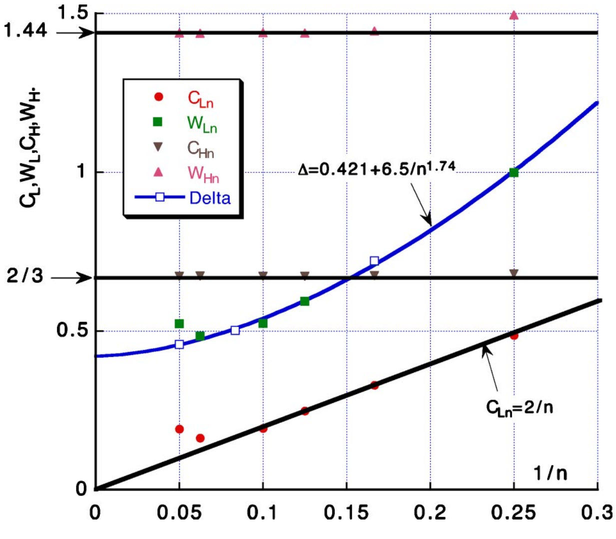

FIG. 4. (Color online) Variation of the parameters C_{Ln}, W_{Ln}, C_{Hn}, and W_{Hn} of the exponentials characterizing the asympotic low and high temperature Arrhenius regimes of $\chi T/(Ng^2\mu_B^2/k)$.

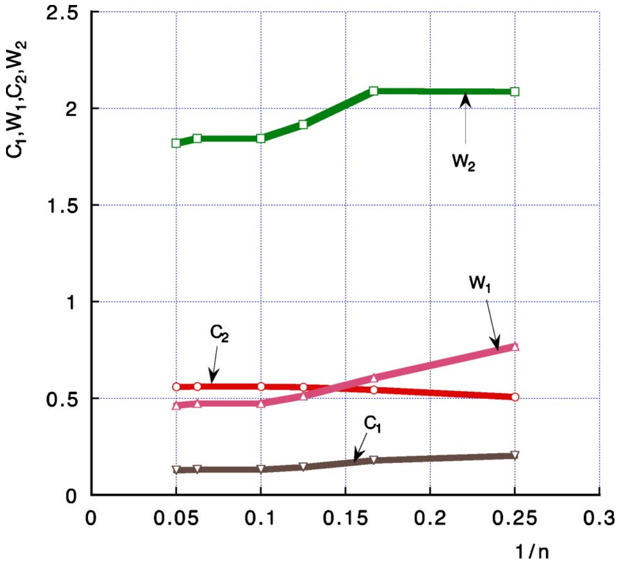

FIG. 5. (Color online) Best values of the parameters C_{1n}, W_{1n}, C_{2n}, and W_{2n}, characterizing the overall temperature dependence of the susceptibility of AF rings of n spins $S=1$ from a two exponential fit.

TABLE II. Results of a two exponential fit of the susceptibility of Y_2BaNiO_5 (from Ref. 8), together with those of $Ni(C_2H_8N_2)_2NO_2ClO_4$, (NENP) along the *a* and *b* axes (see Fig. 6). The data compare to the theoretical values provided $J=300$ K and $g=2.28$ for Y_2BaNiO_5 and 43 K and $g=2.1$ for NENP.

	C_1 (emu.K/mole)	W_1 (K)	C_2 (emu.K/mole)	W_2 (K)	$10^4\chi_0$ (emu/mole)
Y_2BaNiO_5	0.267	138	1.07	534	
NENP-a	0.210	23	0.921	77.7	6.06
NENP-b	0.200	17	0.885	77.4	2.5

$$\chi T = C_{1n} \exp(- W_{1n}/T) + C_{2n} \exp(- W_{2n}/T), \tag{9}$$

whose coefficients (Table I) are relevant for temperature region $T/J > 0.1$. Notice that for $T < 0.1J$, the finite size spectra do not allow a reliable extrapolation of the properties to the thermodynamic limit.

For the Haldane chain ($n \rightarrow \infty$) limit, the best fit parameters are $C_1 = 0.125$, $W_1 = 0.451J$, $C_2 = 0.564$, and $W_2 = 1.793J$.[14] In Ref. 8, the authors have used the above equation to analyze the magnetic data of Y_2BaNiO_5, a typical AF $S=1$ chain system. The corresponding parameters given in Table II agree with our prediction, provided that $J=300$ K which matches with previous findings ($J=285-322$ K).[15]

In this system, the magnetic measurements have been performed down to 10 mK, so that at low enough temperature, the Curie-like paramagnetism of a small number of chain-breaks ends up being the dominant contribution to the susceptibility. Incidentally, in Ref. 8, this contribution is well fitted by an additional term $C_3 \exp(-W_3/T)$. With $W_3 \ll T$ in this case, we are using the fact that Eqs. (5)–(7) have a common high temperature expansion in terms of $1/T$ that goes beyond the Curie–Weiss law: it contains information about the sign and the scale of the interactions. Note that the defects are expected to have very different effects in gapless 1D systems and Haldane chains.[16] In the former, the main effect is to reduce the correlation length: the magnetism in this case amounts to the Curie-like contribution of the finite segments with an odd number of spins. For the Haldane spin-1 chain, by contrast, which is characterized by a finite correlation length at low temperature, the valence-bond-solid (VBS) model suggests a free spin-$\frac{1}{2}$ at each end of the finite segments, giving rise to a staggered susceptibility, but in turn there is no significant change of the gap value.

We have extended the same analysis to the magnetic behavior of $Ni(C_2H_8N_2)_2NO_2ClO_4$ (referred to as NENP), which is the archetype of the Haldane gap systems.[3] The defect-spin contribution is particularly small in this case because, presumably, of the high quality of the single-crystals, and no Curie-like term is needed in the temperature range under consideration. We have therefore introduced a small temperature independent χ_0, which is a contribution depending on axial symmetry. The agreement between theory and experiment is excellent along the crystal axes *a* and *b*, as may be observed in Fig. 6. We need $J=43$ K along both axes to match the paramagnetic Curie–Weiss temperature which is much the same along *a* and *b* axes (see Fig. 6 and Table II). The effect of the known anisotropy of this system is that the Haldane gap, which we deduce from the fit, ranges from $0.53J$ along the *a* axis to $0.40J$ along the *b* axis when we expect $0.451J$ for the isotropic crystal.

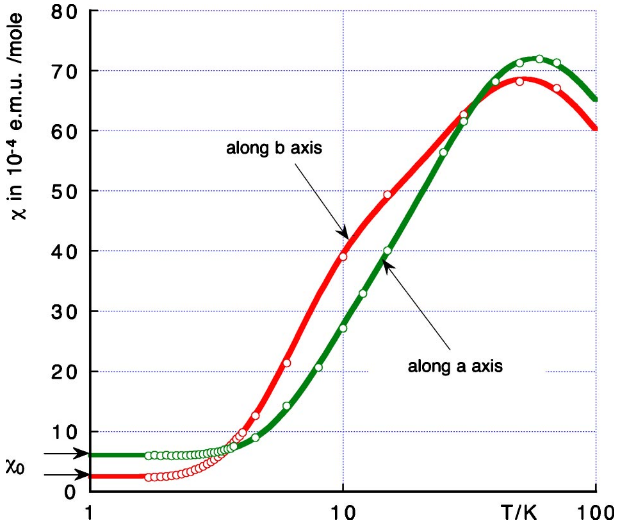

FIG. 6. (Color online) Fit of the magnetic susceptibility data of $Ni(C_2H_8N_2)_2NO_2ClO_4$ (NENP) along two crystallographic axes, giving from the W_2 value, $J=43$ K along all axes. The anisotropy at lower temperature is considered by permitting the Haldane gap to vary from $0.53J$ along the *b* axis to $0.4J$ along the *a* axis instead of $0.447J$ expected for the isotropic case. An additional constant χ_0 has been introduced to account for a residual contribution to the susceptibility that is expected to depend on axial symmetry as observed.

The values of the Curie constants and of the activation energies, deduced for $YBaNiO_5$ and the NENP monocrystal, are listed in Table II.

IV. DISCUSSION

To our knowledge, Eq. (6) is the only expression available to describe the behavior of the Haldane chain in the whole temperature range. At high temperature, everything behaves initially as in any system sitting at a lower critical dimensionality: an exponential solution of the type (6) is found, as in the case of the classical Ising chain with nearest neighbor interactions.

In turn, in the low temperature regime, gapped AF chains are usually described by the expression $\chi = AT^{-1/2} \exp(-\Delta/T)$.[13] The $T^{-1/2}$ factor which follows from field-theory mapping[17] is essentially related to the relativistic magnetic properties in the nanometric scale magnon dispersion,[18] and has further been quantitatively confirmed by Monte Carlo calculations.[19] Actually, it can be pointed out that this expression, which is dominated by the exponential, does not differ significantly from the proposed approach, even for T/J ranging from 0.2 to 0.05, giving $\Delta = 0.409$. Further, it becomes irrelevant at higher temperatures, since little can be inferred from the magnitude of the prefactor, *A*.

In the proposed model, we observe at low temperature the terminal stages of the ordering of a different Ising-like chain, where the Curie constant has been divided by a factor of 5 and the activation energy (now the Haldane gap) by a factor of 3.

Such behavior could be explained by assuming that the N individual moments of the initial chain are not permitted to order completely in a single process where $\xi(T)$ would grow from 0 to ∞ as is described in Sec. II. The initial chain of N spins, rather, is rearranged as a new chain of $N/5$ AF segments of 5 spins each, with an uncompensated spin $S=1$ due to the unbalance of the up and down moments in the correlated state of each segment. The Haldane gap would correspond to the renormalized interaction from segment to segment.

This implies that some sort of dichotomy occurs, at an early stage, whereby a finite length of 5 atomic distances is selected for each segment and the ordering is made in two stages: the former accounts for the short range ordering $0<\xi(T)<\xi_q$ which is responsible for the cohesion of each segment; the latter, $\xi_q<\xi(T)$, with the Curie constant divided by 5, describes the divergence of the correlation between the newly defined segments. The expression (9) displays two simultaneous contributions, accordingly. The segmentation length $\xi_q=5$, which comes out from the ratio of the Curie constants, turns out to be of the same magnitude as the finite correlation length $\xi_q=2JS/\Delta$ predicted for the Haldane chain in the $T=0$ limit.[4] Maybe it is not much of a surprise, after all, if the susceptibility does not vary very much once we reach rings of $n>10$ spins, large enough to contain several segments of size ξ_q. Interestingly, such rings already contain all the elements which are required for describing the physics of the system.

There are many other such cases where the observed experimental evidence can be convincingly described as the superposition of two critical processes in the same temperature range: for example, in ferrimagnetic 1D systems,[10] the minimum of the χT dependence can be well described by two superimposed exponentials, namely ferromagnetic and antiferromagnetic-like contributions, and at the same time a particular length is selected which shows up as a modulation of the ferromagnetic order. Perhaps some general strategy can be elaborated for these systems along the same lines as described here.

V. CONCLUSIONS

We have derived a simple phenomenological expression for the susceptibility $\chi(T)$ of finite AF rings of n Heisenberg spins $S=1$ by applying to the results of exact calculations a method based on physical scaling arguments. On increasing n, the coefficients of this expression rapidly converge to values that do not depend on n anymore. The resulting formula is simple and easy to use: it beautifully fits the experimental magnetic data of $\mathrm{Ni(C_2H_8N_2)_2NO_2ClO_4}$ (NENP) and $\mathrm{Y_2BaNiO_5}$, two archetypes of the Haldane gap systems.

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[1] F. D. M. Haldane, Phys. Lett. A **93**, 464 (1983); Phys. Rev. Lett. **50**, 1153 (1983).

[2] R. Botet and R. J. M. Kolb, Phys. Rev. B **28**, 3914 (1983).

[3] J. P. Renard, M. Verdaguer, L. P. Regnault, W. A. C. Erkelens, J. Rossat-Mignot, and W. G. Stirling, Europhys. Lett. **3**, 945 (1987).

[4] J. P. Renard, L. P. Regnault, and M. Verdaguer, in *Magnetism: Molecules to Materials*, edited by J. S. Miller and M. Drillon (Wiley VCH, Weinheim, 2001), p. 49.

[5] M. M. Nightingale and H. W. Blöte, Phys. Rev. B **33**, 659 (1986); M. Takahashi, *ibid.* **38**, 5188 (1988).

[6] S. R. White, Phys. Rev. Lett. **69**, 2863 (1992); Phys. Rev. B **48**, 10 345 (1993).

[7] J. Souletie, J. Phys. (Paris) **49**, 1211 (1988); Ann. Phys. (Paris) **10**, 69 (1985).

[8] V. Villar, R. Mélin, C. Paulsen, J. Souletie, E. Janod, and C. Payen, Eur. Phys. J. B **25**, 39 (2002).

[9] E. Carré and J. Souletie, J. Magn. Magn. Mater. **72**, 29 (1988); J. Souletie, Y. Tabata, T. Taniguchi, and Y. Miyako, Eur. Phys. J. B **8**, 43 (1999).

[10] M. Drillon, P. Panissod, P. Rabu, J. Souletie, V. Ksenofontov, and P. Gütlich, Phys. Rev. B **65**, 104404 (2002).

[11] S. R. White and D. A. Huse, Phys. Rev. B **48**, 3844 (1993); E. S. Sorensen and I. Affleck, Phys. Rev. Lett. **71**, 1633 (1993).

[12] S. K. Pati, S. Ramasesha, and D. Sen, Phys. Rev. B **55**, 8894 (1997); J. Phys.: Condens. Matter **9**, 8707 (1997).

[13] D. J. Buttrey, J. D. Sullivan, and A. L. Rheingold, J. Solid State Chem. **88**, 291 (1990).

[14] The coefficients for the thermodynamic limit have been deduced by extrapolating the $1/n$ dependence of the C's and W's using third order polynomials.

[15] T. Kennedy, J. Phys.: Condens. Matter **2**, 5737 (1990).

[16] J. Darriet and L. P. Regnault, Solid State Commun. **86**, 409 (1993); R. Saez-Puche, J. M. Coronado, C. L. Otero-Diaz, and J. M. Martin Llorente, J. Solid State Chem. **93**, 461 (1991); J. V. Alvarez, R. Valenti, and A. Zheludev, Phys. Rev. B **65**, 184417 (2002).

[17] I. Affleck, Phys. Rev. B **41**, 6697 (1990).

[18] L. P. Regnault, I. A. Zaliznyak, and S. V. Meshkov, J. Phys.: Condens. Matter **5**, L677 (1993).

[19] Y. J. Kim, M. Greven, U. J. Wiese, and R. J. Birgeneau, Eur. Phys. J. B **4**, 291 (1998).