Thermodynamics of alternating (s,s') chains in the nearest-neighbor Ising-model approximation

J. Curély and R. Georges
Laboratoire de Chimie du Solide, Université de Bordeaux I, 351 Cours de la Libération, 33405 Talence CEDEX, France
(Received 25 February 1992)

The exact solutions of the so-called $(\frac{1}{2},s')_N$ ferrimagnetic *z-z* chain made up of two sublattices have been previously derived by using the transfer-matrix method. In this paper, we generalize this method to $(s,s')_N$ chains, with arbitrary s and s'. We give a general expression for the correlation length ξ and the product $\chi_\parallel T$ in the low-temperature range. We specifically discuss the case of the moment-compensated chain in the low-temperature limit and generalize the results previously obtained. We show that, in this limit, the chain behaves as an assembly of quasi-independent, quasirigid blocks, each with length ξ, and that, in the low-temperature range, the product $\chi_\parallel T$ behaves as ξM^2, with M the thermal-average magnitude of the magnetic moment attributed to the unit cell.

I. INTRODUCTION

In recent years, several theoretical studies on the so-called one-dimensional materials have focused on the thermodynamics of regular exchange-coupled chains.[1-5] Sometimes closed-form, exactly solvable expressions for the thermodynamical functions of interest (specific heat, correlation functions, magnetization, zero-field susceptibility, etc) may be derived for infinite—or finite—regular chains for the case in which the exchange interaction involves spin components parallel or normal to a given axis (*z-z* or planar models)[6-9] or spin operators considered in the classical limit with arbitrary spin dimensionality.[10,11] In a previous paper,[12] we have established general conditions which must be obeyed by the chain Hamiltonian in order to allow an analytical treatment. In particular, we have shown that the *z-z* model is exactly solved when the external field is applied along the z axis. In the other cases, approximate techniques are required to estimate the behavior in the infinite chain limit: Some examples are spin-wave theory,[13] high-temperature series expansions,[14-16] Green's-function approaches,[17] or numerical extrapolation from exact calculations on finite-length chains applied when an isotropic exchange coupling is assumed.[18-22]

The present work has been stimulated by the recent synthesis of bimetallic quasi-one-dimensional complexes $MM'(\text{EDTA})\cdot 6H_2O$ (where EDTA is an abbreviation for ethylenediaminetetracetic acid; M and M' stand for divalent transition metals), the structure of which (from the magnetic point of view) may be represented schematically as infinite zigzag chains[23] of alternating metals $\cdots M$-M'-M-M' $\cdots$. We have focused on the general behavior of quantum or quantum-classical ferrimagnetic chains $(s,s')_N$ described by a *z-z* exchange coupling[24] ($s=\frac{1}{2}$, $s'\geq\frac{1}{2}$). These chains are considered as one-dimensional (1D) systems involving the alternation of two kinds of magnetic moment (which differ by their quantum spin numbers s and s' and/or their Landé factors g and g'). In order to solve this problem, we have used a transfer-matrix method.[25] Among several original results, those concerning the compensation problem appear to be of particular interest since they bring out the interplay between short-range and long-range orders.

In the present paper dealing with ferrimagnetic chains showing *z-z* exchange coupling we generalize the results obtained for the $(\frac{1}{2},s')_N$ chain and consider $(s,s')_N$ chains with arbitrary s and s'. For our present purpose (see below) we may take $s'\geq s$ without loss of generality. Due to the structure of the chain Hamiltonian which will be used, the thermodynamical properties are not modified when s and s' are interchanged, and this will not restrict the range of applicability of the present work. We specifically discuss a bit more the compensation case (no net magnetic moment in the ground state), which also reveals subtle aspects of both short- and long-range orderings. We mainly build up a "map" in the (s,s') plane describing the infinite chain behavior in that case.

II. TRANSFER-MATRIX METHOD AND PARTITION FUNCTION

The transfer-matrix method is well suited for solving *z-z* exchange-coupled chains under an external magnetic field $\mathbf{B}$ applied along the z axis (Fig. 1). Since only the z component of each spin is involved in the Hamiltonian, the corresponding Hamiltonian for the $(2N+1)$-spin chain $s_0s'_0 \cdots s_is'_i \cdots s_N$ may be written

$$\mathcal{H}=\sum_{i=0}^{N-1} H_i^{\text{ex}}+\sum_{i=0}^{N} H_i^{\text{mag}} , \tag{1}$$

with, for a regular chain,

$$H_i^{\text{ex}}=J(s_i^z+s_{i+1}^z)s_i'^z, \quad H_i^{\text{mag}}=-(gs_i^z+g's_i'^z)B . \tag{2}$$

Note that, in the edge contribution, $-g's_N'^zB$ must be dropped. We shall restrict our discussion to antiferrimagnetic $(J>0)$ coupling.

Let $Z_N(B)$ be the partition function for that chain. Let $\mathbf{Z}_N(B)$ be the vector defined as follows: m_N represents the quantized z component of $\mathbf{s}_N$ and is used to define the current state of that spin; among all the states of the chain contributing to the sum $Z_N(B)$, we consider those ones for which the last spin $\mathbf{s}_N$ is in the state described by

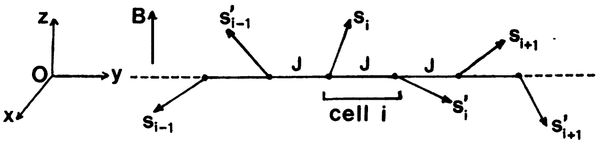

FIG. 1. Structure of a quantum ferrimagnetic $(s,s')_N$ chain.

m_N. The corresponding part of the sum gives the component $Z_N^{m_N}(B)$. As a result, the vector $\mathbf{Z}_N(B)$ has $2s+1$ components, and we have

$$Z_N(B)=\sum_{m_N=-s}^{+s} Z_N^{m_N}(B) \ . \tag{3}$$

We can similarly define the quantities $Z_{N-1}(B)$, $\mathbf{Z}_{N-1}(B)$, and $Z_{N-1}^{m_{N-1}}(B)$ associated to a chain beginning at $\mathbf{s}_0$ and ending at $\mathbf{s}_{N-1}$. It is known that the vectors $\mathbf{Z}_{N-1}(B)$ and $\mathbf{Z}_N(B)$ are related by a matrix operation so that, by repeating this operation along the chain, we can write

$$\mathbf{Z}_N=[\mathcal{T}(B)]^N\mathbf{Z}_0(B) \ , \tag{4}$$

where the so-called transfer matrix $\mathcal{T}(B)$ is a square $(2s+1)\times(2s+1)$ matrix [s has been taken smaller or equal to s' in order to reduce the size of the matrix $\mathcal{T}(B)$]. Its current element appears to be

$$[\mathcal{T}(B)]_{m_i m_{i-1}}=\exp(\beta g m_i B)\frac{\sinh\left[\frac{\beta}{2}(2s'+1)\left[J(m_i+m_{i-1})-\frac{gr}{2s'}B\right]\right]}{\sinh\left[\frac{\beta}{2}\left[J(m_i+m_{i-1})-\frac{gr}{2s'}B\right]\right]} \ , \tag{5}$$

where $r=g's'/gs$ is the ratio of the magnetic-moment magnitudes. Clearly, due to the regularity of the chain, $\mathcal{T}(B)$ appears to be independent of i. Let us label $v_1(B,T)$, $v_2(B,T)$, . . . , $v_{2s+1}(B,T)$, the eigenvalues of the matrix $\mathcal{T}(B)$, ordered in the decreasing modulus order. It is known that, in the infinite chain limit, the whole physics of the extensive parameters is contained in the dependence of the dominant (largest modulus) eigenvalue $v_1(B,T)$ on the relevant conjugated intensive parameters. Specifically, we have for the parallel magnetization $\mathcal{M}_\parallel$ and the zero-field susceptibility $\chi_\parallel$ referred to the unit cell:[25]

$$\mathcal{M}_\parallel=\frac{1}{\beta}\frac{\partial \ln[v_1(B,T)]}{\partial B} \ ,$$
$$\chi_\parallel=\frac{1}{\beta}\left[\frac{1}{v_1(B,T)}\frac{\partial^2 v_1(B,T)}{\partial B^2}\right]_{B=0} \ . \tag{6}$$

In order to get a clearer insight, we shall preferably consider the product $\chi_\parallel T$ normalized to its infinite temperature value (the Curie constant): $(\chi_\parallel T)_n$. We will particularly focus on its low-temperature behavior, more specifically in the compensation case ($r=1$).

The next quantity of significance in the present context is the correlation length ξ which may be defined as follows:

$$\xi=\left[\frac{1}{2}\frac{\sum_{n=0}^{+\infty} n^2|\langle s_0^z s_n^z\rangle|}{\sum_{n=0}^{+\infty}|\langle s_0^z s_n^z\rangle|}\right]^{1/2} \ . \tag{7}$$

Clearly, the correlation length is not an extensive measurable and this will require the knowledge of more than the dominant eigenvalue. Indeed, it is shown (Appendix A) on the basis of the symmetry properties of the matrix $\mathcal{T}(0)$ that, in the low-temperature limit, the correlation length behaves as

$$\xi(T)\sim\frac{1}{1-\left|\frac{v_2(0,T)}{v_1(0,T)}\right|} \quad \text{as } T\to 0 \ . \tag{8}$$

We thus need the low-temperature behavior of the two largest eigenvalues $v_1(0,T)$ and $v_2(0,T)$.

III. THE $(\frac{1}{2},s')_N$ CHAIN

The method described in the preceding section has been used,[26,27] for solving the $(\frac{1}{2},s')_N$ chain problem for both finite and infinite s'. Analytical expressions have been obtained for the correlation length and the parallel susceptibility $\chi_\parallel$ and a brief discussion of their low-temperature behavior has been given. Specifically, near absolute zero, the correlation length behavior is given by

$$\xi(T)\sim\frac{\exp(\beta J s')}{2(2s'+1)} \quad \text{as } T\to 0 \tag{9}$$

and the $(\chi_\parallel T)_n$ product by

$$(\chi_\parallel T)_n\sim\frac{\left[1-r+\frac{r}{s'}\exp(-\beta J)\right]^2\exp(\beta J s')}{(2s'+1)\left[1+\frac{s'+1}{3s'}r^2\right]} \quad \text{as } T\to 0 \ . \tag{10}$$

If the magnetic moments $g/2$ and $g's'$ carried by the

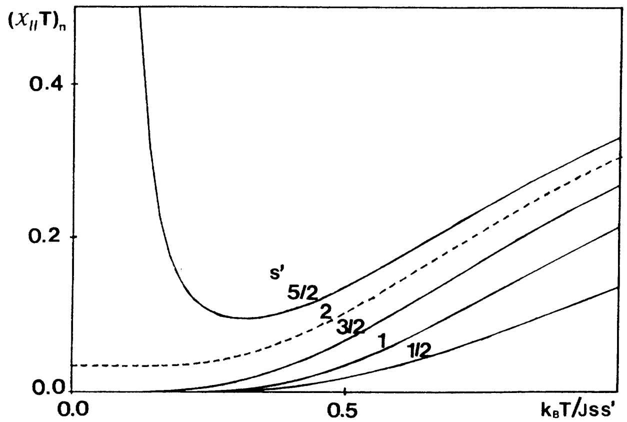

FIG. 2. Thermal variation of the product $(\chi_{\parallel}T)_n$ of a compensated $(\frac{1}{2},s')_N$ chain with z-z type couplings, for various s' values ($J>0$, $g=2$, $r=1$, from Ref. 27).

two sites are unequal ($r\neq 1$), the $(\chi_{\parallel}T)_n$ product diverges as $\exp(\beta Js')$, in the low-temperature limit (like the correlation length). When these moments are equal ($r=1$), the chain has no net moment at absolute zero; some kind of indetermination is expected. It appears that the behavior of the product $(\chi_{\parallel}T)_n$ depends strongly on the spin quantum number s': for $s'<2$, $(\chi_{\parallel}T)_n$ vanishes exponentially as naively expected in such a quasiantiferromagnetic case; however, for $s'>2$, it diverges exponentially. In the intermediate case ($s'=2$), it has a finite limit (Fig. 2).

IV. THE GENERAL $(s,s')_N$ CASE

The problem has been solved by Suzuki *et al.*[8] for $s=s'$ and $g=g'$ but they did not propose a general expression for the product $(\chi_{\parallel}T)_n$: it was observed that this quantity vanishes exponentially at absolute zero. For $s=s'$ and $g\neq g'$, Curély *et al.*[27] ($s=\frac{1}{2}$) and Georges *et al.*[24] ($s=1$) have, respectively, shown on the basis of closed- form expressions that the product $(\chi_{\parallel}T)_n$ diverges exponentially (Table I). In the general case $s'\geq s, g\neq g'$, we now propose a mathematical treatment and give a closed-form expression for the product $(\chi_{\parallel}T)_n$ near absolute zero.

For the nonvanishing field, the matrix $\mathcal{T}(B)$ can be written

$$\mathcal{T}(B)\sim\mathcal{T}(0)+B\,\mathcal{T}'(0)+\frac{B^2}{2}\mathcal{T}''(0)+\cdots \quad \text{as } B\to 0\ , \tag{11}$$

where $\mathcal{T}'(0)$ and $\mathcal{T}''(0)$ are, respectively, the first and second derivatives of $\mathcal{T}(B)$ with respect to B evaluated at $B=0$. The general expression of the current element of these matrices is given in Appendix B [Eqs. (B1)–(B4)]. This allows one immediately to observe that $\mathcal{T}(0)$ and $\mathcal{T}''(0)$ show a symmetry center whereas $\mathcal{T}'(0)$ is antisymmetric. These particular properties are used to transform $\mathcal{T}(B)$ owing to an appropriate unitary transformation described by the matrix $\mathcal{U}$ given in Appendix B [Eqs. (B13)–(B15)]. The new matrix $\tilde{\mathcal{T}}(B)$ has the following forms, depending on the parity of $2s$:

$$s=(2\sigma-1)/2,\sigma\geq 2:\quad \tilde{\mathcal{T}}(B)=\begin{bmatrix}\mathcal{A} & 0\\ 0 & \mathcal{D}\end{bmatrix}+B\begin{bmatrix}0 & \mathcal{B}'\\ \mathcal{C}' & 0\end{bmatrix}+\frac{B^2}{2}\begin{bmatrix}\mathcal{A}'' & 0\\ 0 & \mathcal{D}''\end{bmatrix}+\cdots\ , \tag{12}$$

$$\begin{aligned}s=\sigma,\sigma\geq 2:\quad \tilde{\mathcal{T}}(B)=&\begin{bmatrix} & \mathcal{A} & & 0 & 0\\ t(0,s)\sqrt{2} & \cdots & t(0,1)\sqrt{2} & t(0,0) & 0\\ & 0 & & 0 & \mathcal{D}\end{bmatrix}+B\begin{bmatrix}0 & 0 & & \mathcal{B}' & \\ 0 & 0 & t'(0,s)\sqrt{2} & \cdots & t'(0,1)\sqrt{2}\\ \mathcal{C}' & 0 & & 0 & \end{bmatrix}\\ &+\frac{B^2}{2}\begin{bmatrix} & \mathcal{A}'' & & 0 & 0\\ t''(0,s)\sqrt{2} & \cdots & t''(0,1)\sqrt{2} & t''(0,0) & 0\\ & 0 & & 0 & \mathcal{D}''\end{bmatrix}+\cdots\ ,\end{aligned} \tag{13}$$

TABLE I. Low-temperature behavior of the product $(\chi_{\parallel}T)_n$ for $(s,s')_N$ chains with z-z type coupling.

$r\neq 1$:

$$(\chi_{\parallel}T)_n\sim\frac{(r-1)^2\exp(2\beta Jss')}{(2s'+3-\delta_{s,1/2}-\delta_{s,1})\left[\frac{s+1}{3s}+\frac{s'+1}{3s'}r^2\right]}\quad \text{as } T\to 0,\ \text{with } s'\geq s\ ,$$

$r=1$:

$$(\chi_{\parallel}T)_n\sim\frac{\exp[2\beta Js(s'-2)]}{s'^2(2s'+3-\delta_{s,1/2}-\delta_{s,1})\left[\frac{s+1}{3s}+\frac{s'+1}{3s'}\right]}\quad \text{as } T\to 0\ ,\ \text{with } s'>s\ ,$$

$$(\chi_{\parallel}T)_n\sim\frac{3\exp(-2\beta Js^2)}{2s(s+1)(2s+3-\delta_{s,1/2}-\delta_{s,1})}\quad \text{as } T\to 0\ ,\ \text{with } s'=s\ .$$

where $\mathcal{A}$, $\mathcal{D}$, $\mathcal{B}'$, $\mathcal{C}'$, $\mathcal{A}''$, and $\mathcal{D}''$ are $\sigma\times\sigma$ submatrices given in Appendix B; $t(m,m')$ is the current element of $\mathcal{T}(0)$ and $t'(m,m')$, $t''(m,m')$ are the corresponding first and second derivatives with respect to B evaluated at $B=0$. For $s\le 1$ $(\sigma=1)$ the submatrices reduce to a single element.

For $s=1$, $\mathcal{T}(B)$ is a 3×3 matrix and it remains possible (although a tedious work) to obtain exact expressions for the dominant eigenvalue $v_1(B,T)$ and the related thermodynamical functions. For $s>1$, this generally becomes impossible: it is then necessary, in general, to use a numerical diagonalization process. In the present work, we have focused on the zero-field parallel susceptibility $\chi_\parallel$ determined through a variational process. More specifically, we expand the dominant eigenvalue $v_1(B,T)$ as

$$v_1(B,T)=v_1(0,T)+B\left[\frac{dv_1(B,T)}{dB}\right]_{B=0}+\frac{B^2}{2}\left[\frac{d^2v_1(B,T)}{dB^2}\right]_{B=0}+\cdots . \quad (14)$$

Due to the field-reversal symmetry $[dv_1(B,T)/dB]_{B=0}$ vanishes (as well as all the odd terms). The zero-field eigenvalues $v_1(0,T)$ $v_2(0,T),\ldots,$ $v_{2s+1}(0,T)$ belong to the submatrices $\mathcal{A}$ and $\mathcal{D}$. For $s\le\frac{5}{2}$ $(\sigma\le 3)$, exact expressions remain available. Using a convenient differentiation process,[28] it is possible to express the quantity $[d^2v_1(B,T)/dB^2]_{B=0}$ in terms of the secular polynomial coefficients and their related derivatives with respect to B. For $s>\frac{5}{2}(\sigma>3)$, a numerical diagonalization is unavoidable. However, in the low-temperature range, a perturbation-type calculation may be set on to get the corresponding behaviors of the correlation length and the parallel susceptibility.

As already noted above, near absolute zero, the correlation length ξ is a significant physical parameter; it is related to the ratio $v_2(0,T)/v_1(0,T)$ between the dominant eigenvalues of the $\sigma\times\sigma$ symmetrical submatrices $\mathcal{A}$ and $\mathcal{D}$ (with $\sigma\ge 2$). Actually, it appears (Appendix C) that these eigenvalues are obtained through an ordinary perturbation calculation, which involves only two rows and columns in each independent submatrix. Finally, the low-temperature behavior of the correlation length $\xi(T)$ is given by the expression

$$\xi(T)\sim\frac{\exp(2\beta Jss')}{2(2s'+3-\delta_{s,1/2}-\delta_{s,1})}\quad \text{as } T\to 0\ , \quad (15)$$

which reduces to Eq. (9) for $s=\frac{1}{2}$ ($\delta_{s,u}$ is the Kronecker symbol). The exponential divergence of $\xi(T)$ thus appears to be characteristic of 1D systems involving z-z couplings between spin components.

For the parallel susceptibility $\chi_\parallel$, considered in the low-temperature range, we have developed another perturbation method, based on similar arguments (Appendix D) although the submatrices $\mathcal{B}'$, $\mathcal{C}'$, $\mathcal{A}''$, and $\mathcal{D}''$ are not Hermitian. This method permits to obtain a B expansion for $v_1(B,T)$ [Eq. (D4)] from which we can derive the low-temperature expression [Eq. (D9)] for the product $(\chi_\parallel T)_n$:

$$(\chi_\parallel T)_n\sim\frac{\exp(2\beta Jss')}{(2s'+3-\delta_{s,1/2}-\delta_{s,1})\left[\dfrac{s+1}{3s}+\dfrac{s'+1}{3s'}r^2\right]}\times\left[\left[1-r+\frac{r}{s'}\exp(-2\beta Js)\right]^2-\frac{\exp(-4\beta Js')-\exp(-4\beta Jss')}{s^2}\right]\quad \text{as } T\to 0\ , \quad (16)$$

which generalizes Eq. (10).

In the general case $s'\ne s$, if the magnetic moments gs and $g's'$ carried by the two sites are not equal $(r\ne 1)$, the ground-state magnetic moment of the chain is infinite and the $(\chi_\parallel T)_n$ product diverges at low temperature, like the correlation length ξ, i.e., as $\exp(2\beta Jss')$. When they are equal $(r=1)$, the chain has no net moment near absolute zero. We are dealing with a situation which looks like antiferromagnetism. Actually, as observed for the $(\frac{1}{2},s')_N$ chain, the behavior of the product $(\chi_\parallel T)_n$ is governed by the value of the largest spin quantum number (say s') through the term $\exp[2\beta Js(s'-2)]$: for $s'<2$, $(\chi_\parallel T)_n$ vanishes exponentially as would be expected for a 1D antiferromagnet; on the contrary, for $s'>2$, it diverges exponentially, a rather unexpected behavior. In the intermediate case $(s'=2)$, it has a finite limit.

In the particular case $s=s'$, $g=g'$, the product $(\chi_\parallel T)_n$ vanishes exponentially as $\exp(-2\beta Js^2)$, whatever the common value of s and s'. These results confirm and generalize previous ones dealing with $(\frac{1}{2},s')_N$ chains. They are summarized in Table I and are illustrated by the curves of Figs. 3 and 4. So far, we have just considered spin quantum numbers s and s' such as $s'\ge s$; but, owing to the symmetry of the problem with respect to the interchange of s and s', we can extend these results to all values for s and s': this allows us to build up a map in the whole (s,s') plane, for the low-temperature behavior (Fig. 5) of the product $(\chi_\parallel T)_n$.

In order to interpret the behavior of $(\chi_\parallel T)_n$ in the low-temperature range, we have previously suggested[27] that, in this respect, the chain can be considered as an assembly of independent quasirigid blocks, each one with length ξ (the correlation length). Then, if M is the temperature-dependent magnitude of the magnetic mo-

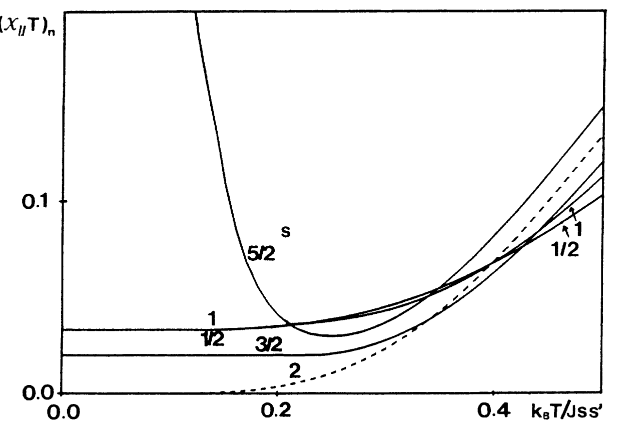

FIG. 3. Thermal variation of the product $(\chi_\parallel T)_n$ of a compensated $(s,2)_N$ chain with z-z-type couplings, for various s values ($J>0$, $g=2$, $r=1$).

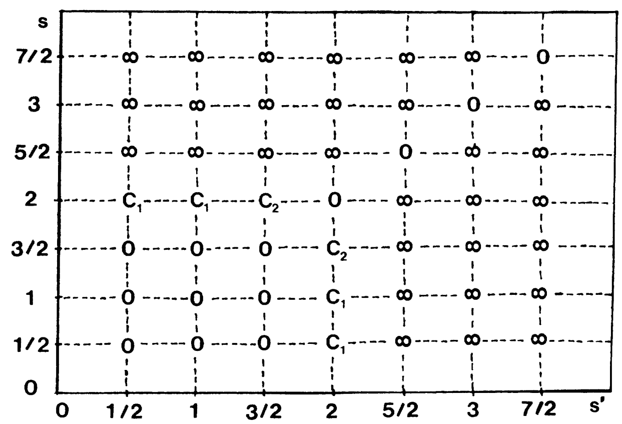

FIG. 5. "Map" in the (s,s') plane of the behavior of the product $(\chi_\parallel T)_n$ at $T=0$ K for a compensated $(s,s')_N$ chain with z-z type couplings ($J>0$, $g=2$, $r=1$); the symbol meaning is 0—the $(\chi_\parallel T)_n$ product vanishes; C—the product shows a finite limit ($C_1=\frac{1}{30}$, $C_2=\frac{1}{42}$); ∞—the product diverges.

ment per unit cell, the product $(\chi_\parallel T)_n$ behaves like ξM^2, with a mathematical indetermination when simultaneously ξ diverges and M vanishes. This model appears to be also relevant for the general $(s,s')_N$ chain. In the noncompensated case ($r\neq 1$), M reaches a finite limit at 0 K and $(\chi_\parallel T)_n$ diverges like ξ, i.e., like $\exp(2\beta Jss')$. In the compensated case ($r=1$), a competition appears between the divergence of ξ and the evanescence of M which is easily shown to vary like $\exp(-2\beta Js)$. The results of Table I show that, in that case too, the low-temperature behavior of $(\chi_\parallel T)_n$ is exactly given by that of ξM^2.

It has been pointed out for the $(\frac{1}{2},s')_N$ chain that, for $s'>2$ and r slightly larger than unity, the mean moment M vanishes at a finite temperature.[27] This is a consequence of the available energy-level density which is larger for spins larger than 2 than for spins $\frac{1}{2}$: as T decreases, the former saturates more slowly than the latter, thus leading to the possibility of exact cancellation if the moment that they carry is the largest one. At that temperature, the susceptibility vanishes: this is the 1D analog of the well-known compensation point met in various 3D ferrimagnets. In the classical limit $s'\rightarrow+\infty$, the same kind of behavior has been predicted but the divergence of ξ and the evanescence of M are expected to depend on the dimensionality d of the space available to the classical spin vector $\mathbf{S}'$.[27] If $d=1$, the behavior of $\mathbf{S}'$ looks like that of a spin $\frac{1}{2}$ and the product $(\chi_\parallel T)_n$ vanishes in the low-temperature limit. If $d=3$, we are dealing with the case $s'>2$ and the product $(\chi_\parallel T)_n$ diverges. When M and T are close to zero, the same compensation point appears because of the continuous character of the classical spin energy-level spectrum. These predictions can be extended to the general $(s,s')_N$ case with $s'\geq 2$ (Fig. 6).

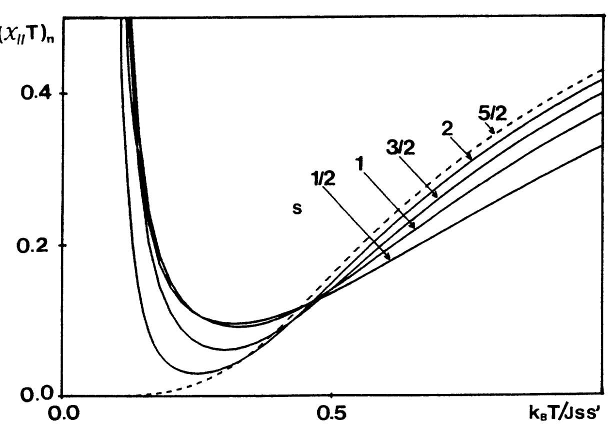

FIG. 4. Thermal variation of the product $(\chi_\parallel T)_n$ of a compensated $(s,\frac{5}{2})_N$ chain with z-z-type couplings, for various s values ($J>0$, $g=2$, $r=1$).

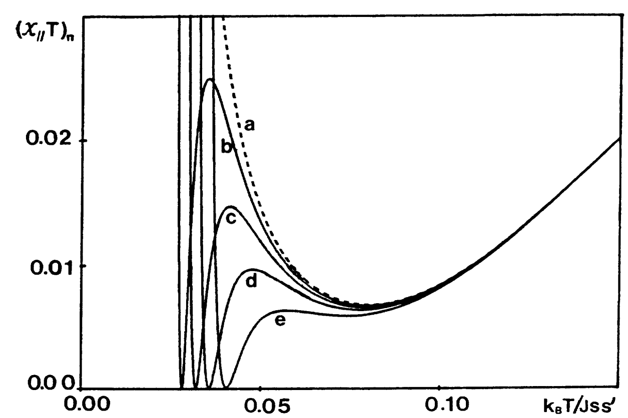

FIG. 6. Thermal variation of the product $(\chi_\parallel T)_n$ of a $(\frac{3}{2},\frac{5}{2})_N$ chain with z-z-type couplings, for various r values near the compensation case ($J>0$, $g=2$; a: $r=1.0000$, b: $r=1.0002$, c: $r=1.0005$, d: $r=1.0010$, e: $r=1.0020$).

V. CONCLUSION

The transfer-matrix method has been applied to the problem of $(s,s')_N$ chains showing z-z couplings between nearest neighbors. Except for the correlation length, the whole physics of the chain is contained in the dominant eigenvalue $v_1(B,T)$ of the matrix $\mathcal{T}(B)$. An analytical expression of $v_1(B,T)$ is available when the smallest of quantum numbers s and s' is lower than $\frac{3}{2}$. For $s \geq \frac{3}{2}$, the secular problem must be solved numerically. However, since we have focused on the parallel susceptibility, it was only necessary to get the zeroth- and second-order terms in the B expansion of $v_1(B,T)$. Up to $s=\frac{5}{2}$, it remains possible to obtain the zero-field eigenvalues and express the second derivative of $v_1(B,T)$ with respect to B by a convenient differentiation of the secular polynomial. For $s>\frac{5}{2}$, a numerical diagonalization is unavoidable. However, in the low-temperature range, the behaviors of the correlation length and of the parallel susceptibility may be derived through a perturbation-type process, whatever the values of s and s'. This allowed us to build up a map, in the (s,s') plane, for describing the behavior of the product $(\chi_{\parallel}T)_n$ for a compensated chain. It appears that this behavior results from a competition between the divergence of the correlation length and the evanescence of the moment magnitude per unit cell. For slightly uncompensated chains, with the largest moment associated with the largest spin, a 1D analog of the well-known compensation point of 3D ferrimagnets is predicted.

APPENDIX A

We want to express the correlation length ξ in order to evaluate its low-temperature behavior. In fact, by using the definition [Eq. (7)], we must calculate the correlation $\langle s_i^z s_j^z \rangle$ that we define as

$$\langle s_i^z s_j^z \rangle = \frac{X}{Z_N(0)} \tag{A1}$$

with

$$X=\sum_{m_0}\cdots\sum_{m_i}\cdots\sum_{m_j}\cdots\sum_{m_N} m_i m_j [\mathcal{T}(0)^{N-j}]_{m_N m_j}[\mathcal{T}(0)^{j-i}]_{m_j m_i}[\mathcal{T}(0)^{i}]_{m_i m_0} Z_0^{m_0}(0) , \tag{A2}$$

where m_i is the z component of the spin $\mathbf{s}_i$; $Z_N(0)$ is deduced from X by substituting 1 to $m_i m_j$. Moreover, we can write

$$\mathcal{T}(0)=\mathcal{S}\mathcal{V}(0,T)\mathcal{S}^{-1} , \tag{A3}$$

where the matrix $\mathcal{V}(0,T)$ is obtained by diagonalizing $\mathcal{T}(0)$ in such a way that the eigenvalues $v_1(0,T)$, $v_2(0,T)$, . . . , $v_{2s+1}(0,T)$ are written in decreasing modulus order since the chain is regular. Equation (A2) can also be written

$$X=\sum_{m_0}\cdots\sum_{m_i}\cdots\sum_{m_j}\cdots\sum_{m_N}\sum_{l}\sum_{l'}\sum_{l''} m_i m_j (\mathcal{S})_{m_N l}[v_l(0,T)]^{N-j}(\mathcal{S}^{-1})_{l m_j}$$
$$\times(\mathcal{S})_{m_j l'}[v_{l'}(0,T)]^{j-i}(\mathcal{S}^{-1})_{l' m_i}(\mathcal{S})_{m_i l''}[v_{l''}(0,T)]^{i}(\mathcal{S}^{-1})_{l'' m_0} Z_0^{m_0}(0) . \tag{A4}$$

Let us suppose that, for finite j-i, the chain length increases to infinity with N-j and i becoming infinite too. In these conditions, for $l\neq 1$ and $l''\neq 1$, the ratios $[v_l(0,T)/v_1(0,T)]^{N-j}$ and $[v_{l''}(0,T)/v_1(0,T)]^{i}$ vanish and we have

$$X=\sum_{m_0}\cdots\sum_{m_i}\cdots\sum_{m_j}\cdots\sum_{m_N}\sum_{l} m_i m_j (\mathcal{S})_{m_N 1}(\mathcal{S}^{-1})_{1 m_j}(\mathcal{S})_{m_j l}[v_l(0,T)]^{j-i}(\mathcal{S}^{-1})_{l m_i}(\mathcal{S})_{m_i 1}(\mathcal{S}^{-1})_{1 m_0} Z_0^{m_0}(0) . \tag{A5}$$

Let us define

$$F_{m_j l}=\sum_{m_{j+1}}\cdots\sum_{m_N}(\mathcal{S})_{m_N 1}(\mathcal{S}^{-1})_{1 m_j}(\mathcal{S})_{m_j l} , \tag{A6}$$

$$G_{l m_i}=\sum_{m_0}\cdots\sum_{m_{i-1}}(\mathcal{S}^{-1})_{l m_i}(\mathcal{S})_{m_i 1}(\mathcal{S}^{-1})_{1 m_0} Z_0^{m_0}(0) . \tag{A7}$$

For j-i much greater than unity, we can only consider the two dominant eigenvalues $v_1(0,T)$ and $v_2(0,T)$ in the l summations present in X. We then get

$$\langle s_i^z s_j^z \rangle = \frac{\displaystyle\sum_{m_i}\sum_{m_j} m_i m_j \left[F_{m_j 1}G_{1 m_i} + \left[\frac{v_2(0,T)}{v_1(0,T)}\right]^{j-i} F_{m_j 2}G_{2 m_i} + \cdots \right]}{\displaystyle\sum_{m_i}\sum_{m_j} \left[F_{m_j 1}G_{1 m_i} + \left[\frac{v_2(0,T)}{v_1(0,T)}\right]^{j-i} F_{m_j 2}G_{2 m_i} + \cdots \right]} . \tag{A8}$$

Due to the symmetry associated to the change of z into $-z$, the elements $(\mathcal{S})_{1m}$ and $(\mathcal{S})_{1-m}$ are equal. Similarly, since in the zero-field limit $\mathcal{T}(0)$ is a symmetrical matrix (and $\mathcal{S}$ consequently a unitary one), the elements $(\mathcal{S}^{-1})_{1m}$ and $(\mathcal{S}^{-1})_{1-m}$ are equal and we can write

$$\sum_m m(\mathcal{S})_{1m}(\mathcal{S}^{-1})_{m1}=0 \ . \tag{A9}$$

As a result, the contribution $m_i m_j F_{m_i 1} G_{1m_i}$ vanishes. Thus, for j-i much greater than unity, Eq. (A8) reduces to

$$|\langle s_i^z s_j^z \rangle| \sim |u|^{j-i}, \quad u=\frac{v_2(0,T)}{v_1(0,T)} \ . \tag{A10}$$

Since $|u|$ is smaller than unity and using Eq. (7) we get immediately

$$\xi^2=\frac{1}{2}\frac{|u|(1+|u|)}{(1-|u|)^2} \ . \tag{A11}$$

In the low-temperature limit $|u|$ is close to unity and the correlation length ξ behaves as

$$\xi(T)\sim\frac{1}{1-\left|\dfrac{v_2(0,T)}{v_1(0,T)}\right|} \quad \text{as } T\to 0 \ . \tag{A12}$$

APPENDIX B

Using Eq. (5) we can easily calculate the elements $d[\mathcal{T}(B)]_{mm'}/dB$ and $d^2[\mathcal{T}(B)]_{mm'}/dB^2$; let us call when $B=0$:

$$t(m,m')=[\mathcal{T}(0)]_{mm'} \tag{B1}$$

and $t'(m,m'), t''(m,m')$ the corresponding expressions of $d[\mathcal{T}(B)]_{mm'}/dB$ and $d^2[\mathcal{T}(B)]_{mm'}/dB^2$ taken for $B=0$; we get

$$t'(m,m')=\beta g s t(m,m')\left[\frac{m}{s}-\frac{r}{2s'}[\tilde{S}\coth(\tilde{S}x)-\coth x]\right] , \tag{B2}$$

$$t''(m,m')=(\beta g s)^2 t(m,m')\left[\left[\frac{m}{s}-\frac{r}{2s'}[\tilde{S}\coth(\tilde{S}x)-\coth x]\right]^2+\left[\frac{r}{2s'}\right]^2\{\tilde{S}^2[1-\coth^2(\tilde{S}x)]-(1-\coth^2 x)\}\right] , \tag{B3}$$

where

$$\tilde{S}=2s'+1, \quad x=\frac{\beta J}{2}(m+m') \ . \tag{B4}$$

These matrix elements show the following symmetry properties:

$$t(-m,-m')=t(m,m'), \quad t'(-m,-m')=-t'(m,m'), \quad t''(-m,-m')=t''(m,m') \ . \tag{B5}$$

According to Eq. (11) we can expand $\mathcal{T}(B)$ in the vanishing field limit; as the size of $\mathcal{T}(B)$ is $(2s+1)\times(2s+1)$ we must distinguish the integer and noninteger s cases.

Let us consider, for instance, the case $s=\frac{3}{2}$. Owing to Eqs. (B1)–(B5) we have

$$\mathcal{T}(B)=\begin{bmatrix} t(3/2,3/2) & t(3/2,1/2) & t(1/2,1/2) & t(1/2,-1/2) \\ t(3/2,1/2) & t(1/2,1/2) & t(1/2,-1/2) & t(1/2,1/2) \\ t(1/2,1/2) & t(1/2,-1/2) & t(1/2,1/2) & t(3/2,1/2) \\ t(1/2,-1/2) & t(1/2,1/2) & t(3/2,1/2) & t(3/2,3/2) \end{bmatrix}$$

$$+B\begin{bmatrix} t'(3/2,3/2) & t'(3/2,1/2) & t'(3/2,-1/2) & t'(3/2,-3/2) \\ t'(1/2,3/2) & t'(1/2,1/2) & t'(1/2,-1/2) & t'(1/2,-3/2) \\ -t'(1/2,-3/2) & -t'(1/2,-1/2) & -t'(1/2,1/2) & -t'(1/2,3/2) \\ -t'(3/2,-3/2) & -t'(3/2,-1/2) & -t'(3/2,1/2) & -t'(3/2,3/2) \end{bmatrix}$$

$$+\frac{B^2}{2}\begin{bmatrix} t''(3/2,3/2) & t''(3/2,1/2) & t''(3/2,-3/2) & t''(3/2,-3/2) \\ t''(1/2,3/2) & t''(1/2,1/2) & t''(1/2,-3/2) & t''(1/2,-3/2) \\ t''(1/2,-3/2) & t''(1/2,-1/2) & t''(1/2,3/2) & t''(1/2,3/2) \\ t''(3/2,-3/2) & t''(3/2,-1/2) & t''(3/2,1/2) & t''(3/2,3/2) \end{bmatrix}+\cdots \ . \tag{B6}$$

Applying the unitary transformation

$$\tilde{\mathcal{T}}(B)=\mathcal{U}\mathcal{T}(B)\mathcal{U}^{-1} , \tag{B7}$$

defined by the matrix $\mathcal{U}$:

$$\mathcal{U}=\mathcal{U}^{-1}=\frac{1}{\sqrt{2}}\begin{bmatrix}1&0&0&1\\0&1&1&0\\0&1&-1&0\\1&0&0&-1\end{bmatrix}, \tag{B8}$$

we obtain

$$\tilde{\mathcal{T}}(B)=\begin{bmatrix}\mathcal{A}&0\\0&\mathcal{D}\end{bmatrix}+B\begin{bmatrix}0&\mathcal{B}'\\\mathcal{C}'&0\end{bmatrix}+\frac{B^2}{2}\begin{bmatrix}\mathcal{A}''&0\\0&\mathcal{D}''\end{bmatrix}+\cdots, \tag{B9}$$

with

$$\mathcal{A}=\begin{bmatrix}t(3/2,3/2)+t(1/2,-1/2) & t(3/2,1/2)+t(1/2,1/2)\\ t(3/2,1/2)+t(1/2,1/2) & t(1/2,1/2)+t(1/2,-1/2)\end{bmatrix},$$

$$\mathcal{D}=\begin{bmatrix}t(1/2,1/2)-t(1/2,-1/2) & t(3/2,1/2)-t(1/2,1/2)\\ t(3/2,1/2)-t(1/2,1/2) & t(3/2,3/2)-t(1/2,-1/2)\end{bmatrix}, \tag{B10}$$

$$\mathcal{B}'=\begin{bmatrix}t'(3/2,1/2)-t'(3/2,-1/2) & t'(3/2,3/2)-t'(3/2,-3/2)\\ t'(1/2,1/2)-t'(1/2,-1/2) & t'(1/2,3/2)-t'(1/2,-3/2)\end{bmatrix},$$

$$\mathcal{C}'=\begin{bmatrix}t'(1/2,3/2)+t'(1/2,-3/2) & t'(1/2,1/2)+t'(1/2,-1/2)\\ t'(3/2,3/2)+t'(3/2,-3/2) & t'(3/2,1/2)+t'(3/2,-1/2)\end{bmatrix}, \tag{B11}$$

$$\mathcal{A}''=\begin{bmatrix}t''(3/2,3/2)+t''(3/2,-3/2) & t''(3/2,1/2)+t''(3/2,-1/2)\\ t''(1/2,3/2)+t''(1/2,-3/2) & t''(1/2,1/2)+t''(1/2,-1/2)\end{bmatrix},$$

$$\mathcal{D}''=\begin{bmatrix}t''(1/2,1/2)-t''(1/2,-1/2) & t''(1/2,3/2)-t''(1/2,-3/2)\\ t''(3/2,1/2)-t''(3/2,-1/2) & t''(3/2,3/2)-t''(3/2,-3/2)\end{bmatrix}. \tag{B12}$$

This matricial structure extends to all half integers $s=(2\sigma-1)/2$ values, with $\sigma\geq 2$, and $\mathcal{A}$, $\mathcal{D}$, $\mathcal{B}'$, $\mathcal{C}'$, $\mathcal{A}''$, and $\mathcal{D}''$ are also $\sigma\times\sigma$ submatrices. The general form of the transformation matrix $\mathcal{U}$ is

$$\mathcal{U}=\begin{bmatrix}\frac{1}{\sqrt{2}}\imath & \frac{1}{\sqrt{2}}\mathcal{J}\\ \frac{1}{\sqrt{2}}\mathcal{J} & -\frac{1}{\sqrt{2}}\imath\end{bmatrix}, \tag{B13}$$

where $\imath$ and $\mathcal{J}$ are also $\sigma\times\sigma$ submatrices defined by

$$(\imath)_{ij}=\delta_{ij},\quad (\mathcal{J})_{ij}=\delta_{i\sigma+1-j}. \tag{B14}$$

For integer s values ($s=\sigma$), the structure is slightly different; the matrix $\mathcal{U}$ must be written

$$\mathcal{U}=\begin{bmatrix}\frac{1}{\sqrt{2}}\imath & 0 & \frac{1}{\sqrt{2}}\mathcal{J}\\ 0&1&0\\ \frac{1}{\sqrt{2}}\mathcal{J} & 0 & -\frac{1}{\sqrt{2}}\imath\end{bmatrix}, \tag{B15}$$

where $\imath$ and $\mathcal{J}$ are defined as above. The structure of $\tilde{\mathcal{T}}(B)$ is now

$$\tilde{\mathcal{T}}(B)=\begin{bmatrix}\mathcal{A} & 0 & 0\\ \sqrt{2}t(0,s)\cdots\sqrt{2}t(0,1) & t(0,0) & 0\\ 0 & 0 & \mathcal{D}\end{bmatrix}+B\begin{bmatrix}0&0&\mathcal{B}'\\0&0&\sqrt{2}t'(0,s)\cdots\sqrt{2}t'(0,1)\\ \mathcal{C}'&0&0\end{bmatrix}$$

$$+\frac{B^2}{2}\begin{bmatrix}\mathcal{A}'' & 0 & 0\\ \sqrt{2}t''(0,s)\cdots\sqrt{2}t''(0,1) & t''(0,0) & 0\\ 0&0&\mathcal{D}''\end{bmatrix}, \tag{B16}$$

where $\mathcal{A}$, $\mathcal{D}$, $\mathcal{B}'$, $\mathcal{C}'$, $\mathcal{A}''$, and $\mathcal{D}''$ are also $\sigma\times\sigma$ submatrices.

APPENDIX C

In order to evaluate the correlation length [Eq. (8)] in the low-temperature range, we must calculate the first two dominant eigenvalues $v_1(0,T)$, $v_2(0,T)$ of the matrix $\tilde{\mathcal{T}}(0)$. At low temperature, the leading terms of the current matrix element $t(m,m')$ are

$$t(m,m')\sim\exp(K|m+m'|)[1+\exp(-k|m+m'|)+\exp(-2k|m+m'|)+\cdots] \text{ as } T\to 0 \tag{C1}$$

with

$$K=\beta Js', \quad k=\beta J . \tag{C2}$$

Let us notice that $t(m,-m)$ is equal to $\tilde{\mathcal{S}}$ [cf. Eq. (B4)].

It appears that the $s=\frac{1}{2}$ and $s=1$ cases require specific treatments. For $s=\frac{1}{2}$, we have

$$\tilde{\mathcal{T}}(0)=\begin{bmatrix} t(1/2,1/2)+t(1/2,-1/2) & 0 \\ 0 & t(1/2,1/2)-t(1/2,-1/2)\end{bmatrix}, \tag{C3}$$

from which we deduce immediately

$$\xi(T)\sim\frac{\exp(\beta Js')}{2(2s'+1)}, \quad s=\tfrac{1}{2} \text{ as } T\to 0 . \tag{C4}$$

For $s=1$, we get

$$\tilde{\mathcal{T}}(0)=\begin{bmatrix} t(1,1)+t(0,0) & 0 & 0 \\ t(0,1)\sqrt{2} & t(0,0) & 0 \\ 0 & 0 & t(1,1)-t(0,0)\end{bmatrix} . \tag{C5}$$

The first and third diagonal terms are the dominant eigenvalues and we get similarly

$$\xi(T)\sim\frac{\exp(2\beta Js')}{2(2s'+1)} \text{ as } T\to 0, \text{ with } s=1 . \tag{C6}$$

In the general case ($s>1$), the matrix $\tilde{\mathcal{T}}(0)$ has a general form which depends on the parity of $2s$. We have

$$\tilde{\mathcal{T}}(0)=\begin{bmatrix} \mathcal{A} & 0 \\ 0 & \mathcal{D}\end{bmatrix}, \quad 2s \text{ odd } [s=(2\sigma-1)/2, \sigma\geq 2] , \tag{C7}$$

$$\tilde{\mathcal{T}}(0)=\begin{bmatrix} \mathcal{A} & 0 & 0 \\ t(0,s)\sqrt{2}\cdots t(0,1)\sqrt{2} & t(0,0) & 0 \\ 0 & 0 & \mathcal{D}\end{bmatrix}, \quad 2s \text{ even } (s=\sigma, \sigma\geq 2) . \tag{C8}$$

In both cases, $\mathcal{A}$ and $\mathcal{D}$ are the $\sigma\times\sigma$ symmetrical submatrices defined by

$$\mathcal{A}=\begin{bmatrix} t(s,s)+t(s,-s) & t(s-1,s)+t(s-1,-s) & \cdots \\ t(s-1,s)+t(s-1,-s) & t(s-1,s-1)+t(s-1,1-s) & \cdots \\ & & \cdots \\ \vdots & \vdots & \cdots \end{bmatrix},$$

$$\mathcal{D}=\begin{bmatrix} \cdots & & \\ \cdots & \vdots & \vdots \\ \cdots & t(s-1,s-1)-t(s-1,1-s) & t(s-1,s)-t(s-1,-s) \\ \cdots & t(s-1,s)-t(s-1,-s) & t(s,s)-t(s,-s)\end{bmatrix} . \tag{C9}$$

In the low-temperature limit we have

$$t(s,s)\pm t(s,-s)\sim\exp(2Ks)[f_1(k)\pm\tilde{S}\exp(-2Ks)] \text{ as } T\to 0 , \tag{C10}$$

$$t(s-1,s-1)\pm t(s-1,1-s)\sim\exp(2Ks)[\exp(-2K)f_2(k)\pm\tilde{S}\exp(-2Ks)] \text{ as } T\to 0 , \tag{C11}$$

$$t(s-1,s)\pm t(s-1,-s)\sim\exp(2Ks)\{\exp(-K)f_3(k)\pm\exp[-K(2s-1)]f_4(k)\} \text{ as } T\to 0 , \tag{C12}$$

where $f_1(k), f_2(k), f_3(k), f_4(k)$ may be expanded as

$$f_1(k)=1+\exp(-2ks)+\exp(-4ks)+\cdots , \tag{C13}$$

$$f_2(k)=1+\exp[-2k(s-1)]+\exp[-4k(s-1)]+\cdots , \quad \text{(C14)}$$

$$f_3(k)=1+\exp[-k(2s-1)]+\exp[-2k(2s-1)]+\cdots , \quad \text{(C15)}$$

$$f_4(k)=1+\exp(-k)+\exp(-2k)+\cdots . \quad \text{(C16)}$$

At low temperature, it appears that the remaining matrix elements introduce nonsignificant contributions to the correlation length ξ. As a result, the general form of the two dominant eigenvalues can be written

$$v(0,T)\sim\exp(2Ks)\left[f_1(k)\pm\tilde{S}\exp(-2Ks)+\frac{\{\exp(-K)f_3(k)\pm\exp[-K(2s-1)]f_4(k)\}^2}{f_1(k)-\exp(-2K)f_2(k)}\right] \text{ as } T\to 0 , \quad \text{(C17)}$$

where the upper (lower) sign refers to $v_1(0,T)$ [$v_2(0,T)$]; we thus deduce for the correlation length the expression

$$\xi(T)\sim\frac{\exp(2\beta Jss')}{2(2s'+3)} \text{ as } T\to 0, \text{ with } s>1 . \quad \text{(C18)}$$

Owing to Eqs. (C4) and (C6), we can generalize the expression of $\xi(T)$ given in the above equation:

$$\xi(T)\sim\frac{\exp(2\beta Jss')}{2(2s'+3-\delta_{s,1/2}-\delta_{s,1})} \text{ as } T\to 0 . \quad \text{(C19)}$$

APPENDIX D

In this appendix we evaluate the parallel susceptibility of an $(s,s')_N$ chain ($s'\geq s$) and more specifically the product $\chi_\parallel T$. For the particular cases $s=\frac{1}{2}$ and $s=1$, the calculation is straightforward and we get easily

$$s=\tfrac{1}{2}:\ \chi_\parallel T\sim\left[\frac{g}{2}\right]^2\left[1-r+\frac{r}{s'}\exp(-\beta J)\right]^2\frac{\exp(\beta Js')}{2s'+1} \text{ as } T\to 0 , \quad \text{(D1)}$$

$$s=1:\ \chi_\parallel T\sim g^2\left[1-r+\frac{r}{s'}\exp(-2\beta J)\right]^2\frac{\exp(2\beta Js')}{2s'+1} \text{ as } T\to 0 . \quad \text{(D2)}$$

In the general case ($s>1$) we are compelled to use a perturbation method; as pointed out in Appendix C, $\tilde{\mathcal{T}}(0)$ is a symmetrical matrix but $\tilde{\mathcal{T}}'(0)$ and $\tilde{\mathcal{T}}''(0)$ are not. We assume that we can expand $v_1(B,T)$ as a power series in B:

$$v_1(B,T)=v_1(0,T)+\frac{B^2}{2}\left[\frac{d^2v_1(B,T)}{dB^2}\right]_{B=0}+\cdots . \quad \text{(D3)}$$

The odd terms vanish because of field reversal symmetry. Despite the non-Hermiticity of $\tilde{\mathcal{T}}'(0)$ and $\tilde{\mathcal{T}}''(0)$ but thanks to the Hermiticity of $\tilde{\mathcal{T}}(0)$, it appears that the second-order B perturbation can be handled, in a rather similar way to the well-known Hermitian case. The first step consists in calculating the matrices $\tilde{\mathcal{T}}(0)$, $\tilde{\mathcal{T}}'(0)$, and $\tilde{\mathcal{T}}''(0)$ in the eigenbasis of $\tilde{\mathcal{T}}(0)$, thus giving $\tilde{\tilde{\mathcal{T}}}(0)$, $\tilde{\tilde{\mathcal{T}}}'(0)$, and $\tilde{\tilde{\mathcal{T}}}''(0)$. Assuming that the largest eigenvalue of $\tilde{\tilde{\mathcal{T}}}''(0)$ corresponds to the first eigenvector, the B dependence of $v_1(B,T)$ may be written

$$v_1=\tilde{\tilde{\mathcal{T}}}_{11}+\frac{B^2}{2}\left[\tilde{\tilde{\mathcal{T}}}''_{11}+2\sum_{i\neq 1}\frac{\tilde{\tilde{\mathcal{T}}}'_{1i}\tilde{\tilde{\mathcal{T}}}'_{i1}}{v_1(0,T)-v_i(0,T)}\right]+\cdots , \quad \text{(D4)}$$

where the B and T variables have been dropped [because of the non-Hermiticity of $\tilde{\tilde{\mathcal{T}}}'(0)$, the product $\tilde{\tilde{\mathcal{T}}}'_{1i}\tilde{\tilde{\mathcal{T}}}'_{i1}$ cannot be replaced by $(\tilde{\tilde{\mathcal{T}}}'_{1i})^2$]. The resulting expression of the parallel susceptibility may be written

$$\chi_\parallel=\frac{1}{\beta v_1(0,T)}\left[\tilde{\tilde{\mathcal{T}}}''_{11}+2\sum_{i\neq 1}\frac{\tilde{\tilde{\mathcal{T}}}'_{1i}\tilde{\tilde{\mathcal{T}}}'_{i1}}{v_1(0,T)-v_i(0,T)}\right] . \quad \text{(D5)}$$

Let us now turn to the evaluation of the various terms involved in Eq. (D5) in the low-temperature range. This necessitates a work similar to that achieved in Appendix C. It is easily shown that, in this limit, the term $\tilde{\tilde{\mathcal{T}}}''_{11}$ can be ignored as well as the terms $\tilde{\tilde{\mathcal{T}}}'_{1i}\tilde{\tilde{\mathcal{T}}}'_{i1}$ with $i>2$. Finally Eq. (D6) reduces to

$$\chi_\parallel\sim\frac{2}{\beta}\frac{\tilde{\tilde{\mathcal{T}}}'_{12}\tilde{\tilde{\mathcal{T}}}'_{21}}{(v_1(0,T))^2}\frac{1}{1-\left|\dfrac{v_2(0,T)}{v_1(0,T)}\right|} \text{ as } T\to 0 , \quad \text{(D6)}$$

where the last fraction is merely the correlation length $\xi(T)$ given by Eq. (A12) and where $\tilde{\tilde{\mathcal{T}}}'_{12}\tilde{\tilde{\mathcal{T}}}'_{21}$ is given by

$$\tilde{\tilde{\mathcal{T}}}'_{12}\tilde{\tilde{\mathcal{T}}}'_{21}\sim[t'(s,s)]^2-\exp(-2K)\{[t'(s,s-1)-t'(s-1,s)]^2-[t'(s,1-s)-t'(s-1,-s)]^2\} \text{ as } T\to 0 . \tag{D7}$$

Using the low-temperature expansion of $t'(m,m')$

$$t'(m,m')\sim\beta gst(m,m')\left[\frac{m}{s}-r+\frac{r}{s'}\exp(-k|m+m'|)\right] \text{ as } T\to 0 \tag{D8}$$

and those ones of $t(m,m')$ and $\xi(T)$ [Eqs. (C1) and (C19)], the product $\chi_{\|}T$ may finally be written

$$\chi_{\|}T\sim(gs)^2\left[\left[1-r+\frac{r}{s'}\exp(-2\beta Js)\right]^2-\frac{\exp(-4\beta Js')-\exp(-4\beta Jss')}{s^2}\right]\frac{\exp(2\beta Jss')}{2s'+3-\delta_{s,1/2}-\delta_{s,1}} \text{ as } T\to 0 . \tag{D9}$$

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