Local-anisotropy and spin-nature effects in ferrimagnetic Ising chains: New compensation phenomena

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We propose a general treatment for solving ferrimagnetic chains, made up of two spin sublattices (s,S), by assuming a Z-Z exchange coupling between nearest neighbors. Exact expressions of the susceptibility will be derived for $s=\frac{1}{2}$ spins alternating either with classical moments or with arbitrary S quantum spins. In the first case, the dimensionality of the space available to classical spins will be taken into account for describing the magnetic behavior. New specific effects will be discussed when the sublattice magnetizations nearly or exactly compensate one another. In particular, the occurrence of a compensation temperature, corresponding to exact cancellation of the opposite magnetizations, as in three-dimensional ferrimagnets, will be revealed. This manifestation will be shown to depend drastically on the spin multiplicities and on the ratio between magnetic moments $2GS/g$.

I. INTRODUCTION

Extensive reports on one-dimensional spin systems arise on the one hand from the large collection of quasi-one-dimensional materials synthesized in the last few years, and on the other hand from their ability to be solved exactly in many nontrivial cases. Thus, thermodynamic functions of interest (specific heat, correlation functions, magnetization, zero-field susceptibility, etc.) have been derived for regular chains when the exchange interaction involves spin components parallel or normal to a given axis (Z-Z or X-Y models).[1-4] Also, large spin systems have been studied using approximate techniques; for instance, high-temperature series expansions or the finite-string method.[5-8] Although appearing at first glance to be of purely academic interest, the classical limit (infinite spin) with isotropic interactions may give some insight into the behavior of systems which would otherwise be intractable.[9,10]

Stimulated by the recent synthesis of new bimetallic quasi-one-dimensional complexes, MM' (EDTA)-6H_2O (EDTA is ethylenediamine tetra-acetic acid), the structure of which may be schematized as infinite zig-zag chains of alternating metals M-M'-M-M'...,[11] we have focused for a time on the general behavior of ferrimagnetic chains $(S_1,S_2)_N$ described by Heisenberg or Z-Z exchange coupling.[12-14]

Among various results, those concerning the compensation problem seem to be the most original ones. Considering, for instance, the fully isotropic quantum chain (Heisenberg coupling, no local anisotropy), it has been shown that the magnetic moments g_1S_1 and g_2S_2 carried by the two kinds of sites must be unequal for the chain to exhibit a quasiantiferromagnetic behavior. In the case of two alternating ($S_1=\frac{1}{2}$, $S_2=1$) spins, the ratio g_1/g_2 must take a value very close to $\frac{8}{3}$ instead of the expected value of 2 (Ref. 12). This clearly results from the Heisenberg nature of the coupling and must be related to the so-called zero-motion spin reduction which is relatively more efficient for smaller spin quantum numbers. However, this must render us cautious when trying to relate the behavior of the ferrimagnetic chain to that of conventional three-dimensional ferrimagnets.

The present paper deals with ferrimagnetic chains showing Z-Z exchange coupling, especially from the viewpoint of the compensation problem that is the condition for quasi-antiferromagnetic behavior in the low-temperature limit. In Sec. II we shall establish that within the conditions which allow for using a transfer-matrix method,[15] it is possible in a number of cases to reduce to 2×2 the size of the matrices of interest, thus allowing an easy determination of the largest eigenvalue in terms of matrix elements. In Sec. III A, we shall consider a ferrimagnetic chain with a quantum spin $\frac{1}{2}$ alternating with a classical one of amplitude S, and a nearest-neighbor Z-Z interaction. Three different situations will be examined: (i) where the classical spins are submitted to an infinite anisotropy which forces them to align along the z direction, (ii) where the classical spins are subject to an infinite anisotropy which favors the x-z plane (containing the coupling axis), and (iii) where the classical spins are free to rotate in the full space (the finite anisotropy problem will be considered in a forthcoming paper). These situations will be characterized by the dimensionality $d=1$, 2, or 3 of the space available to the classical spins. In each case, we shall give expressions for the zero-field susceptibilities $\chi_u^{(d)}$ along the principal directions. They will be used for discussing the very-low-

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temperature behavior and the conditions for the divergence of the $\chi_u^{(d)}T$ product at absolute zero (Secs. III B and III C). Also, the one-dimensional behavior of the so-called compensation point, which is observed in a variety of three-dimensional ferrimagnets, will be described (Sec. III D). Section IV will be devoted to the study of another kind of ferrimagnetic chain. The classical vectors of amplitude S will be replaced by quantum spins with spin quantum number S. They will be subject to no local anisotropy, and will be coupled to the neighboring $\frac{1}{2}$ quantum spins by the same Z-Z interaction as in the preceding section. General expressions for the zero-field susceptibility along the coupling axis will be given. Furthermore, it will be shown that the $\frac{1}{2}$- and infinite-S cases coincide with the $d=1$ and $d=3$ problems of Sec. III, thus providing us with two distinct paths between these extreme systems characterized by quite different behaviors.

II. GENERAL CONSIDERATIONS

Let us consider a finite-length ferrimagnetic chain $(S_0,s_1,S_1,s_2\cdots S_{N-1},s_N,S_N)$ with spin quantum numbers $S,s,S\ldots$, respectively. Assuming nearest-neighbor exchange coupling only, we can write the Hamiltonian for the whole chain:

$$\mathscr{H}=\sum_{i=0}^{N} H_i \tag{1}$$

with

$$H_i=H_i^{(1)}(\mathbf{S}_{i-1},\mathbf{s}_i)+H_i^{(2)}(\mathbf{s}_i)+H_i^{(3)}(\mathbf{s}_i,\mathbf{S}_i)+H_i^{(4)}(\mathbf{S}_i)\ . \tag{2}$$

The contributions $H_i^{(2)}$ and $H_i^{(4)}$ are single-spin energy terms, involving local anisotropy as well as magnetostatic coupling. The terms $H_i^{(1)}$ and $H_i^{(3)}$ deal with the exchange part of the energy. The condition for the transfer-matrix method to operate in the present problem is that all the commutators $[H_i,H_j]$ vanish. This reduces to the conditions

$$[H_i^{(3)}(\mathbf{s}_i,\mathbf{S}_i)+H_i^{(4)}(\mathbf{S}_i),H_{i+1}^{(1)}(\mathbf{S}_i,\mathbf{s}_{i+1})]=0\ . \tag{3}$$

One can set a variety of situations for which these conditions are verified. The main two ones are (i) where the exchange coupling and the single-ion Hamiltonian $H_i^{(4)}$ involve the same unique component of the vector operator $\mathbf{S}_i$ (say, S_i^z, with Z-Z exchange coupling, Z-uniaxial anisotropy, and the external magnetic field $\mathbf{B}$ applied along $\hat{\mathbf{z}}$), and (ii) where the spin quantum number S is large enough for $[S_i^x,S_i^y]$ to be negligible compared to $S_i^xS_i^y$ (classical spin approximation). When the condition (3) is fulfilled the partition function Z_N for the $2N+1$ spin chain may be written

$$Z_N=\mathrm{Tr}\left[\prod_{i=0}^{N}\exp(-\beta H_i)\right],\quad \beta=1/k_BT\ . \tag{4}$$

Let us now introduce a function $U(\mathbf{S}_i)$ as an operator whose eigenvalues $u_1,u_2,\ldots,u_n,\ldots$ are nondegenerate and exhibit a symmetrical distribution characterized by the even function $\rho(u)$. It is then possible to replace any summation over a complete basis describing the states of $\mathbf{S}_i$ by an integration:

$$\sum_{\mathbf{S}_i}\cdots=\int du\,\rho(u)\cdots\ . \tag{5}$$

Now, the trace (4) may be computed by first summing over the quantum states of the spins $\mathbf{s}_i$. This leads to the quantities

$$\phi_i(u_{i-1},u_i)=\sum_{\sigma_i}\langle\sigma_i\,|\exp(-\beta H_i)\,|\,\sigma_i\rangle\ . \tag{6}$$

Suppose now that this function may be expressed as a bilinear expression in terms of even and odd functions of its arguments:

$$\phi_i(u_{i-1},u_i)=\sum_{\epsilon_i=\pm1}\sum_{\eta_i=\pm1}K_i^{\epsilon_i}(u_{i-1})L_{i,\epsilon_i}^{\eta_i}(u_i)\ , \tag{7}$$

where the values $+1$ and -1 of the superscripts mean even and odd, respectively. It is then straightforward to show that the effective partition function for a pair inserted in the infinite chain is merely the largest (essentially positive) eigenvalue of the matrix

$$(\mathscr{C})=\begin{bmatrix}F_{n,1}^{1} & F_{n,-1}^{1}\\ F_{n,1}^{-1} & F_{n,-1}^{-1}\end{bmatrix}\ , \tag{8}$$

where

$$F_{n,\epsilon}^{\epsilon'}=\int du\,\rho(u)L_{n,\epsilon}^{\epsilon'}(u)K_{n+1}^{\epsilon'}(u)\ . \tag{9}$$

Since the eigenvalue problem for 2×2 matrices is solved exactly, the decomposition (7) allows us to obtain analytical expressions for the partition function and the correlated thermodynamic properties. The general formulation may now be applied to the derivation of magnetic properties of various ferrimagnetic chains with Z-Z nearest-neighbor exchange coupling.

III. QUANTUM-CLASSICAL $(\frac{1}{2}\text{-}S)_N$ CHAIN WITH Z-Z INTERACTION

A. Spin Hamiltonian

In this section, the spins $\mathbf{s}_i$ stand for $\frac{1}{2}$ quantum spin operators. As for the spins $\mathbf{S}_i$, we shall consider them as classical vectors with amplitude S. Furthermore, we assume a Z-Z nearest-neighbor coupling with exchange constant J. Thus,

$$H_i^{(1)}=-JS_{i-1}^z s_i^z,\quad H_i^{(3)}=-Js_i^zS_i^z\ . \tag{10}$$

Clearly, the most interesting behavior is expected for an antiferromagnetic coupling between nearest neighbors, as is currently observed in three-dimensional ferrimagnets. Thus, the exchange parameter J will henceforth be restricted to strictly negative values. Moreover, the classical spins will be subject to various conditions. Namely, we shall consider three cases: (i) where they are constrained to lie along the z axis (exchange coupling axis) (ii) where they are restricted to lie within the x-y plane, and (iii) where they are free to orient themselves along any direction of their space. These situations will be designated by the dimension $d=1$, 2, or 3 of the available space. Let us now designate by B the amplitude of the magnetic field applied to the chain, and by g and G the Landé factors for the $\mathbf{s}_i$ and $\mathbf{S}_i$ spins, respectively. We shall use the following notation:

$$a=-\beta JS,\quad b=\beta g\mu_B B/2\ ,$$
$$u_i=S_i^z/S,\quad r=2GS/g\ , \tag{11}$$

where μ_B is the Bohr magneton and r represents the ratio of the magnetic moments carried by the two kinds of spins. The operator $U(\mathbf{S}_i)$ has been chosen to be S_i^z/S. In the present case its eigenvalues are distributed among the interval $(-1,+1)$ with a distribution function reflecting the dimensionality of the available space. Specifically, $d=1$:

$$\rho(u)=\tfrac{1}{2}[\delta(u+1)+\delta(u-1)]\ , \tag{12}$$

$d=2$:

$$\rho(u)=\frac{1}{\pi}(1-u^2)^{-1/2}\ , \tag{13}$$

$d=3$:

$$\rho(u)=\tfrac{1}{2}\ . \tag{14}$$

$\delta(x-a)$ is the so-called Dirac distribution centered at a.

The detailed calculation of the partition function now depends on the direction of the magnetic field $\mathbf{B}$. We shall consider successively the cases where $\mathbf{B}$ is applied along $\hat{\mathbf{z}}$, and normal to $\hat{\mathbf{z}}$. The corresponding susceptibilities for vanishing $\mathbf{B}$ will be noted $\chi_z^{(d)}$ in the first case and $\chi_x^{(d)}$ or $\chi_y^{(d)}$ in the second one. The superscript (d) refers to the dimensionality of the space available to the classical spin vectors.

B. Applied magnetic field along $\hat{\mathbf{z}}$

The influence of the external field is expressed by

$$H_i^{(2)}=-g\mu_B s_i^z B,\quad H_i^{(4)}=-G\mu_B S u_i B\ . \tag{15}$$

Inserting (10) and (15) into (6) and summing over $s_i^z=\pm\frac{1}{2}$, we get

$$\phi_i(u_{i-1},u_i)=2\exp(rbu_i)\cosh\left[b-\frac{a}{2}(u_{i-1}+u_i)\right]\ . \tag{16}$$

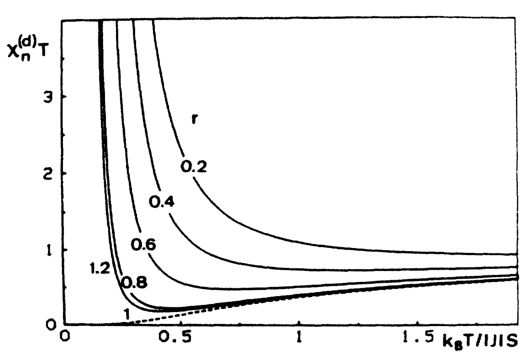

FIG. 1. Magnetic behavior of the quantum-classical $(\frac{1}{2}\text{-}S)_N$ chain for some significant values of $r=2GS/g$; the dimensionality of the classical spin sublattice is assumed to be $d=1$. The susceptibility is plotted in reduced units (see text).

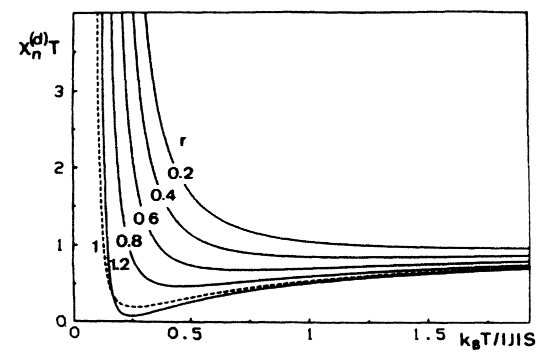

FIG. 2. Magnetic behavior of the quantum-classical $(\frac{1}{2}\text{-}S)_N$ chain for $d=2$ and some significant values of r. The curve corresponding to the compensation value $r=1$ is shown by the dashed line.

This expression clearly obeys the condition (7), and after a little algebra, we obtain

$$F_{i,+1}^{+1}=2(J_1\cosh b+J_3\sinh b)\ , \tag{17}$$

$$F_{i,+1}^{-1}=2(J_3\cosh b+J_2\sinh b)\ , \tag{18}$$

$$F_{i,-1}^{+1}=2(J_3\cosh b+J_1\sinh b)\ , \tag{19}$$

$$F_{i,-1}^{-1}=2(J_2\cosh b+J_3\sinh b)\ , \tag{20}$$

with

$$J_1=\int_{-1}^{+1}du\,\rho(u)\cosh^2(au)\cosh(rbu)\ , \tag{21}$$

$$J_2=\int_{-1}^{+1}du\,\rho(u)\sinh^2(au)\cosh(rbu)\ , \tag{22}$$

$$J_3=\int_{-1}^{+1}du\,\rho(u)\cosh(au)\sinh(au)\sinh(rbu)\ , \tag{23}$$

with ρ depending on d [see (12) to (14)].

J_1,J_2,J_3 may be given analytical expressions in terms of modified Bessel functions of the first kind. Then, the effective partition function and the derivatives of interest may be determined. The main results are given in Table I and illustrated by the curves of Figs. 1 to 3. These figures actually show the thermal variation of the quantity $\chi_n^{(d)}T$, defined as the product $\chi_z^{(d)}T$ normalized to unity in the high-temperature range [the normalizing factor is $k_B/(g\mu_B)^2(1+r^2/d)$] whatever r and d may be, thus allowing an easy comparison of the various situations. It appears immediately that the most interesting features are

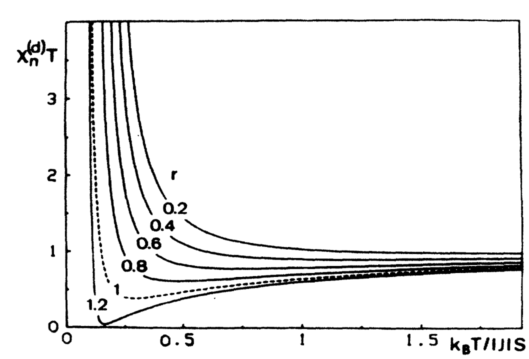

FIG. 3. Magnetic behavior of the quantum-classical $(\frac{1}{2}\text{-}S)_N$ chain for $d=3$ and some significant values of r.

TABLE I. Expressions for the susceptibility of quantum-classical $(\frac{1}{2}\text{-}S)_N$ chains for various situations of local anisotropy on classical spins. The I's are modified Bessel functions of the first kind with argument a. We put $\delta=d/2-1$ and $\Lambda_d=(\pi/2)^{2\delta^2}a^{-\delta}$, with $a=-\beta JS$.

Arbitrary d	$\chi_z^{(d)}=\beta\left[\frac{g\mu_B}{2}\right]^2\left[\frac{r^2}{d}\left[1+\frac{(d-1)\Lambda_d I_{\delta+2}+d(\Lambda_d I_{\delta+1})^2}{1+\Lambda_d I_\delta}\right]-2r\Lambda_d I_{\delta+1}+\Lambda_d I_\delta\right]$
$\lim_{\beta\to 0}\beta^{-1}\chi_z^{(d)}$	$\left[\frac{g\mu_B}{2}\right]^2\left[1+\frac{r^2}{d}\right]$
$d=1$	$\chi_z^{(1)}=\beta\left[\frac{g\mu_B}{2}\right]^2(r^2\cosh a-2r\sinh a+\cosh a)$
$d=2$	$\chi_z^{(2)}=\beta\left[\frac{g\mu_B}{2}\right]^2\left[r^2\frac{1+I_0+I_2+2I_1^2}{2(1+I_0)}-2rI_1+I_0\right]$
$d=3$	$\chi_z^{(3)}=\beta\left[\frac{g\mu_B}{2}\right]^2\left[r^2\frac{3a(a^2+2)\sinh a-6a(a+\sinh a)\cosh a+3(a^2+1)\sinh^2 a+a^2(a^2+3)}{3a^3(a+\sinh a)}-2r\frac{a\cosh a-\sinh a}{a^2}+\frac{\sinh a}{a}\right]$

to be expected for r values not too far from unity.

At very high temperature, the magnetic moments behave nearly independently and the product $\chi_z^{(d)}T$ is merely the sum of the Curie constants on both kinds of sites. As the temperature is lowered the nearest-neighbor correlation increases. Due to the antiferromagnetic nature of the coupling this results in an effective reduction of the magnetic moment to be attributed to a pair of neighboring spins. Meanwhile, the correlation length ξ increases. For r values significantly far from unity, the moment reduction is relatively weak, the increase of ξ dominates, and $\chi_z^{(d)}T$ increases uniformly as T is lowered. If r is closer to unity, the relative moment reduction is more drastic and at first dominates; we thus observe a decrease of $\chi_z^{(d)}T$ which passes through a minimum and diverges in the very-low-temperature limit except, eventually, when r strictly equals unity. As can be seen for the $r=1$ curves of Figs. 1 to 3, the very-low-temperature behavior of the susceptibility depends strongly on the dimensionality of the space available to the classical spins when dealing with compensated sublattices. This can be understood by the analysis of the correlation length. Let us define the mean amplitude $\mathcal{M}$ of the magnetic moment one can attribute to a pair of sites (magnetic cell). So far as we are concerned with the magnetic susceptibility, we can approximate the chain as an assembly of independent quasirigid blocks, each one with length ξ thus carrying a magnetic moment $\mathcal{M}\xi/a$ (a being the magnetic cell parameter). The magnetic susceptibility related to a unit cell appears then to be given by

$$\chi_z^{(d)}=A\beta\xi\mathcal{M}^2\ , \tag{24}$$

where A is a temperature-independent factor. For r different from unity, clearly $\mathcal{M}$ reaches a finite value at absolute zero and $\chi_z^{(d)}T$ diverges like ξ. For $r=1$, we must look more carefully at the exact behavior of ξ and $\mathcal{M}$. It is easily shown (see Appendix A) that ξ diverges according to

$$\xi\simeq\beta^{(1-d)/2}\exp(-\beta JS)\ . \tag{25}$$

As for the magnetic moment being attributed to the magnetic cell, we have for $r=1$

$$\mathcal{M}=g\mu_B\langle\,|s_i^z-S_i^z/2S\,|\,\rangle\ , \tag{26}$$

where $|\cdots|$ refers to the absolute value and $\langle\cdots\rangle$ to the thermodynamical mean value. For $d=2$ and $d=3$, the energy-level spectrum for the classical spins is continuous, whereas it is discrete for the quantum ones. Near absolute zero we are allowed to neglect the fluctuations of s_i^z compared to those of S_i^z, and we may write

$$\mathcal{M}=\tfrac{1}{2}g\mu_B\langle 1-2s_i^zS_i^z/S\rangle\ . \tag{27}$$

$\mathcal{M}$ thus appears to be proportional to that part of the exchange energy E_J which vanishes at absolute zero, and is easily deduced from the zero-field partition function

$$\mathcal{M}\simeq 1-\frac{\Lambda_d I_{\delta+1}}{1+\Lambda_d I_\delta} \tag{28}$$

(for the meanings of δ, Λ_d, and I_μ, see Table I), which is simply proportional to T near absolute zero. Finally, we obtain

$$\chi_z^{(d)}\simeq\beta^{-(d-3)/2}\exp(-\beta JS)\quad(d=2,3)\ , \tag{29}$$

in complete agreement with the exact expressions of Table I. For $d=1$, the classical spins exhibit a discrete-level spectrum, and $\mathcal{M}$ now vanishes exponentially:

$$\mathcal{M}\simeq\exp(\beta JS)\ . \tag{30}$$

Due to this much more drastic decrease, the $\chi_z^{(1)}T$ product appears now to vanish at absolute zero,

$$\chi_z^{(1)}T \simeq \exp(\beta JS) \ . \tag{31}$$

This discussion clearly indicates that the low-temperature behavior of the susceptibility $\chi_z^{(d)}$ results from a sharp competition between long-range correlations, tending to increase the number of magnetic cells which move almost as a whole, and short-range correlations, which tend to reduce the magnetic moment on each cell. Owing to the detailed circumstances one observes various behaviors as described in the present section. Further examples will be given in the next one.

C. Applied magnetic field normal to $\hat{z}$

We now consider the susceptibilities $\chi_x^{(d)}$ and $\chi_y^{(d)}$ corresponding to a vanishing field applied along $\hat{x}$ or $\hat{y}$. Since the related spin components are not coupled by the exchange Hamiltonian, these susceptibilities can be expressed as

$$\chi_v^{(d)} = \chi_{v,q}^{(d)} + r^2 \chi_{v,c}^{(d)} \quad (v = x,y) \ , \tag{32}$$

where $\chi_{v,q}^{(d)}$ and $\chi_{v,c}^{(d)}$ characterize the response to an external field applied along $\hat{v}$ of a unique quantum or classical $(r=1)$ spin, inserted into the chain. The derivation of these expressions is given in Appendices B and C, with some extra details in Appendix D. The results are gathered in Table II and illustrated by the normalized curves of Fig. 4. Some features exhibited by these curves may need some short comments. As expected, the classical contribution (dashed line) vanishes for $d=1$, while for $d=2$ and $d=3$, it decreases linearly at very low temperatures from the initial value $\chi_n^{(d)}J=1$. This results from the linear decrease which initially affects the mean-square Z component of the classical moments (and their moduli) as a consequence of the continuous character of the distribution $\rho(v)$. This effect is twice as effective for $d=3$ as for $d=2$; as a direct consequence, the exchange field on the quantum spins decreases linearly. Since, in this temperature range, these align almost perfectly along the local field, this decrease is the leading effect and the quantum contribution to the susceptibility increases (twice as fast for $d=3$ as for $d=2$). No such effect is to be expected for $d=1$. The upward departure from the initial behavior occurs at higher temperature because the quantum spins, in their turn, no longer align along the local field. The maxima and the decrease observed at intermediate and high temperatures are trivial features.

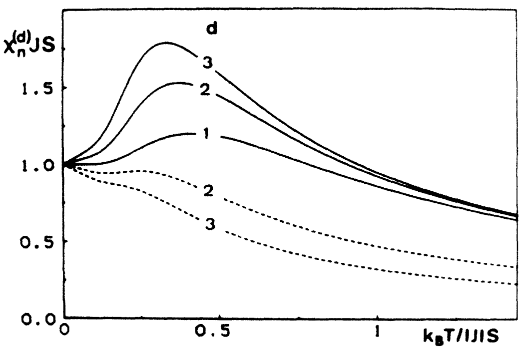

FIG. 4. Thermal variations of the normal susceptibilities (field applied along $\hat{x}$ or $\hat{y}$) of the quantum (solid lines) and classical (dashed lines) sublattices for $d=1$, 2, and 3. We emphasize that $X_{v,c}^{(1)}J$ vanishes at all temperatures.

D. Compensation point

We come now to the question whether a "compensation point" may be expected in some one-dimensional ferrimagnets. In the three-dimensional case the exact cancellation, at Θ_c, of the opposite sublattice magnetizations occurs because of their quite different thermal variations. For instance, in some rare-earth garnets, the transition-metal ions occupy the sublattice A and the rare-earth ones the sublattice B, with a strong (positive) n_{AA} molecular field coefficient, whereas n_{AB} is weaker and negative and n_{BB} may be neglected. As the garnet is cooled down, the

TABLE II. Expressions for the normal susceptibility of quantum-classical $(\frac{1}{2}\text{-}S)_N$ chains for various situations of local anisotropy on classical spins. δ, I_v, and Λ_d are defined in Table I.

$$\chi_v^{(d)} = \chi_{v,q}^{(d)} + r^2 \chi_{v,c}^{(d)} \quad (v = x,y)$$

$$\chi_{v,c}^{(d)} = \beta k_v^{(d)} \left[\frac{g\mu_B}{2} \right]^2 \frac{J_1(b=0) - \frac{1}{4}\left[\left[\frac{\partial^2 J_1}{\partial a^2} \right]_{b=0} + \frac{2}{d} \right]}{J_1(b=0)} \quad (v = x,y)$$

$$\chi_{x,Q}^{(d)} = \chi_{y,Q}^{(d)} = \beta \left[\frac{g\mu_B}{2} \right]^2 (J_4/J_1^2)(b=0)$$

$$J_1(b=0) = \tfrac{1}{2}(1 + \Lambda_d I_\delta)$$

$$J_4 = \frac{1}{2a} a^\delta \Lambda_d \sum_{n=0}^{\infty} \frac{a^{2n+1}}{2n+1} \left[\frac{1}{\sqrt{\pi}} \frac{(n+\delta-\frac{1}{2})!}{n!(n+\delta)!(n+2\delta)!} + \frac{R_{\delta,n}}{2^{2\delta}\pi(2n)!} \right]$$

$$R_{-1/2,n} = n+1, \quad R_{0,n} = \pi, \quad R_{1/2,n} = \frac{4}{n+1} \sum_{l=0}^{n} (2l+1)^{-1}$$

transition-metal ions first order ferromagnetically, weakly polarizing the rare-earth sublattice. As the temperature decreases again, the rare-earth magnetization increases up to saturation in the opposite direction to the transition-metal sublattice. Exact compensation occurs at an intermediate temperature if the B sublattice carries a larger moment than does the A one.

With the present picture of a ferrimagnetic chain, no long-range order has to be expected. Moreover, one cannot easily imagine an intrasublattice (next-nearest-neighbor) exchange coupling that would significantly dominate the intersublattice (nearest-neighbor) one. However, the curves of Fig. 5, drawn for $d=3$ and r values slightly larger than unity, reveal an unexpected behavior for $\chi_z^{(d)}T$. On heating from absolute zero, instead of regularly decreasing down to the ordinary minimum, the product $\chi_z^{(3)}T$ exhibits, at an intermediate temperature, an extra minimum which is surprisingly close to zero. A similar behavior is observed under the same conditions for $d=2$. The reason for that behavior is that, because of the continuous character, already mentioned, of their energy-level spectrum, the magnetic moments carried by the classical spins align less efficiently than the quantum ones along the exchange field. Since for r larger than unity they reach a larger value, there is an exact cancellation temperature at which $\mathcal{M}$ vanishes. The different thermal behavior which can result from the difference between n_{AA} and n_{BB} in the garnets is, in the present case, a consequence of the difference between $H_i^{(2)}$ and $H_i^{(4)}$, the single-site parts of the Hamiltonian, that involve the nature of the spins and the local anisotropy. The susceptibility cannot exactly vanish at the minimum, because at finite temperature there is always some amount of fluctuation on the components s_i^z and S_i^z. The conditions for such a behavior to be observed are, thus, (i) that the competing magnetic moments cancel one another (i.e, $r>1$) and (ii) that this cancellation occurs at a temperature at which the spin component fluctuations are negligible and the correlation length ξ very large. This necessitates that r be slightly larger than unity. It is worth noticing that these conditions, which seem to be necessary for the occurrence of a compensation point in the one-dimensional ferrimagnet, should extend to the two- and three-dimensional systems. From a practical point of view such a behavior might be expected whenever a unique cation occupies two sites exhibiting different orbital contributions to the magnetic moments and strongly different alignment tendencies along the local exchange field.

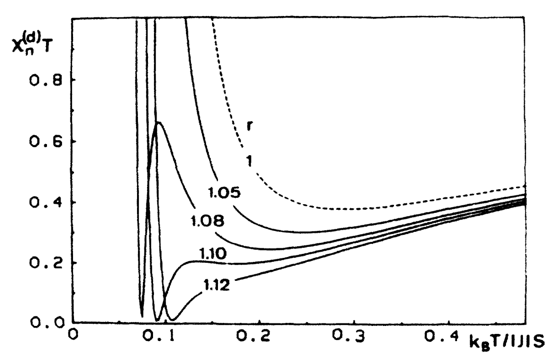

FIG. 5. Magnetic behavior of the quantum-classical $(\frac{1}{2}\text{-}S)_N$ chain for $d=3$ and r slightly larger than unity, showing compensation phenomena.

IV. FERRIMAGNETIC $(\frac{1}{2},S)_N$ QUANTUM CHAIN WITH Z-Z INTERACTION

We consider a ferrimagnetic chain $(S_0,s_1,S_1,\ldots s_N,S_N)$ as described in Sec. II but, contrary to Sec. III, we assume now that $\mathbf{S}_i$ refers to a quantum spin operator defined by S. Because of this modification, it is no longer possible to get exact expressions for the susceptibilities in vanishing fields applied normally to the coupling z axis. Thus, we shall only be concerned with the so-called parallel susceptibility which we shall designate by $\chi_z^{[S]}$, where the superscript refers to the quantum number of $\mathbf{S}_i$ spins. The area examined in the present section is twice that of the previous ones. The infinite-S limit exactly coincides with the $d=3$ case previously encountered. Furthermore, at least from the parallel susceptibility point of view, the $S=\frac{1}{2}$ problem is strictly equivalent to the $d=1$ case. Thus, one expects a drastic change in the low-temperature behavior of the product $\chi_z^{[S]}T$ as S decreases from infinity down to $\frac{1}{2}$. And since it cannot occur in a continuous way, one has to point out the turning conditions.

Clearly, the considerations of Sec. II and Appendix A do prevail, since only s_i^z and S_i^z operators are involved in the chain Hamiltonian. Moreover, all the relations (10) to (23) remain formally valid except for the distribution function $\rho(u)$ which takes the general form

$$\rho(u)=(2S+1)^{-1}\sum_{\sigma=-S}^{S}\delta(u-\sigma/S)\ . \tag{33}$$

The calculations are straightforward and we easily get

$$\chi_z^{[S]}=\beta(Y_1+r^2Y_2)\ , \tag{34}$$

where

$$Y_1=\frac{P}{Q}\mu_B^2\frac{\sinh(Qa/2)}{\sinh(Pa/2)}\times\left[1+\frac{r}{2S}\left[\cosh(Pa/2)-\frac{Q}{P}\coth(Qa/2)\right]\right]^2\ , \tag{35}$$

$$Y_2=\{1+[Q\sinh(Pa/2)]/[P\sinh(Qa/2)]\}^{-1}\ , \tag{36}$$

and

$$P=1/2S,\quad Q=(2S+1)/2S\ . \tag{37}$$

Clearly Y_2 vanishes at absolute zero for any nonvanishing S. A necessary condition for $\chi_z^{[S]}$ not to diverge is thus that Y_1 vanishes too, that merely implies $r=1$, which is nothing but the ordinary condition for the magnetic-moment equality. Now the question arises whether this conditions is a sufficient one. A detailed examination of expression (35) shows that $\chi_z^{[S]}$ behaves according to the law

$$\chi_z^{[S]}\simeq\beta\exp[-\beta J(S-2)] \tag{38}$$

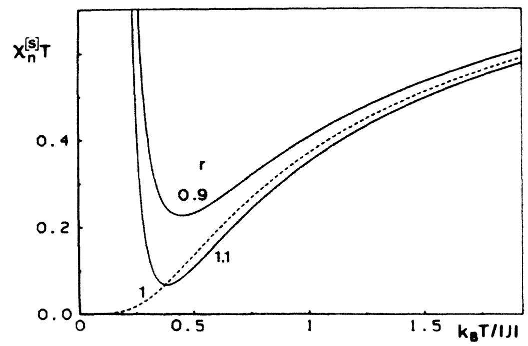

FIG. 6. Magnetic behavior of the ferrimagnetic chain $(\frac{1}{2}-\frac{3}{2})_N$ for some significant values of r.

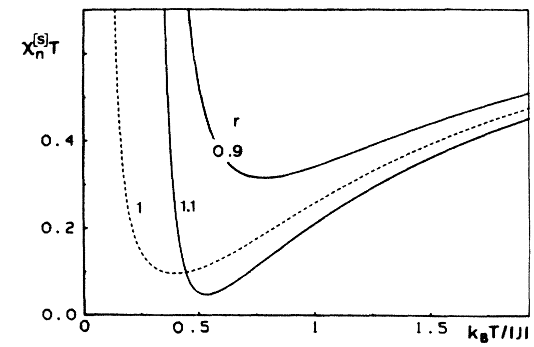

FIG. 8. Magnetic behavior of the ferrimagnetic chain $(\frac{1}{2}-\frac{5}{2})_N$ for some significant values of r.

in the low-temperature range. As a result, the product $\chi_z^{[S]}T$ diverges for S greater than 2, in agreement with the $d=3$-type behavior expected for large S. Conversely, this product vanishes for S smaller than 2, in agreement with the $d=1$ behavior, while for the turning value $S=2$ it exhibits a finite limit. The curves of Figs. 6, 7, and 8 illustrate these behaviors for $S=\frac{3}{2}$, 2, and $\frac{5}{2}$. We have reported the thermal variation of the quantity $\chi_n^{[S]}T$ defined as the product $\chi_z^{[S]}T$ normalized to unity in the high-temperature range; the normalizing factor is in this case

$$k_B/(g\mu_B)^2[1+r^2S(S+1)/3] \ .$$

As in Sec. III B one can discuss the $r=1$ case in terms of correlation length ξ and magnetic moment per magnetic cell, which are easily shown to vary like $\exp(-\beta JS)$ and $\exp(+\beta J)$ in the very-low-temperature limit, thus leading, after (24), to expression (38).

Furthermore, the curves of Fig. 9 drawn for $S=\frac{5}{2}$ and r slightly larger than unity clearly show the existence of a compensation point. This is a consequence of the available energy-level density which is larger for spin $\frac{5}{2}$ than for spin $\frac{1}{2}$. When T decreases, the former saturates more slowly than the latter, thus leading to the possibility of exact cancelling if the moment they carry is the largest one.

V. CONCLUSION

In the present paper we have set a general formulation for solving in an easy way the statistical problem of a large class of one-dimensional ferrimagnets. The transfer matrices are built up *a priori* in their diagonal form, which makes it easier to study a large variety of problems. Besides those which are investigated in this paper we believe that, for instance, the question of random coupling[16,17] should be an interesting field of application for this method. We have derived exact expressions for the principal magnetic susceptibilities for a Z-Z coupled chain showing classical spin vectors alternating with $s=\frac{1}{2}$ quantum ones. The case where the classical spins are replaced by quantum operators with arbitrary spin quantum number S has also been treated from the parallel susceptibility point of view. The very-low-temperature behavior of the principal susceptibilities has been discussed in some detail. In particular, it has been shown that new specific effects have to be expected whenever the sublattice magnetizations nearly or exactly compensate one another. However, their detailed manifestations should strongly depend on the nature of the spins and on the local anisotropies. We believe that such predictions should induce experimental works on nearly compensated one-dimensional ferrimagnets, elaborated for instance by introducing the same magnetic cation into two significantly different host sites arranged to constitute an alternating chain.

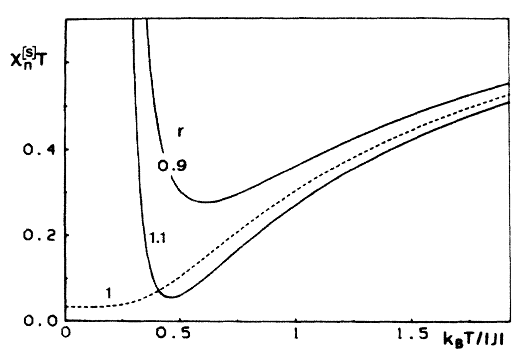

FIG. 7. Magnetic behavior of the ferrimagnetic chain $(\frac{1}{2}-2)_N$ for some significant values of r.

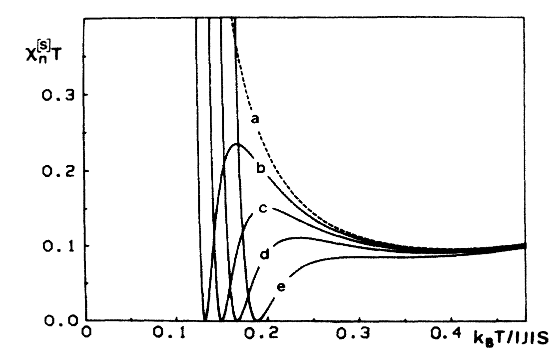

FIG. 9. Magnetic behavior of the ferrimagnetic chain $(\frac{1}{2}-\frac{5}{2})_N$ for values of r equal to or slightly larger than unity. Curve a: $r=1$, curve b: $r=1.0002$, curve c: $r=1.0005$, curve d: $r=1.001$, and curve e: $r=1.002$.

APPENDIX A

Because of the nature of the Hamiltonians $H_i^{(1)}$ and $H_i^{(3)}$ for the ferrimagnetic chains studied in Secs. III and IV, and whenever no external field is applied, $\phi_i(u_{i-1},u_i)$ remains unchanged when one modifies simultaneously the signs of u_{i-1} and u_i. Thus, ϕ_i reduces to

$$\tilde{\phi}_i(u_{i-1},u_i)=\sum_{\epsilon=\pm 1} K_i^{\epsilon}(u_{i-1})L_{i,\epsilon}^{\epsilon}(u_i) \quad \text{(A1)}$$

and $(\mathscr{C}_i)$ to

$$(\tilde{\mathscr{C}}_i)=\begin{bmatrix} F_{i-1,+1}^{+1} & 0 \\ 0 & F_{i-1,-1}^{-1}\end{bmatrix} . \quad \text{(A2)}$$

When the chain exhibits a translational invariance, all the matrices are similar, and the i indices may be dropped. Now, we introduce the field V_m coupled to u_m only. The extra term $-V_m u_m$ adds to $H_m^{(4)}(\mathbf{S}_m)$ and, to first order in βV_m, $\tilde{\phi}_m$ becomes

$$\hat{\phi}_m(u_{m-1},u_m)=\phi(u_{m-1},u_m) +\beta V_m \sum_{\epsilon} u_m \tilde{K}^{\epsilon}(u_{m-1})\tilde{L}_{\epsilon}^{\epsilon}(u_m) . \quad \text{(A3)}$$

Hence, $(\tilde{\mathscr{C}}_{m+1})$ is changed from $(\tilde{\mathscr{C}})$ to

$$(\hat{\mathscr{C}}_{m+1})=(\tilde{\mathscr{C}})+\beta V_m \begin{bmatrix} 0 & f_{-1}^{+1} \\ f_{+1}^{-1} & 0\end{bmatrix} , \quad \text{(A4)}$$

with

$$f_{\epsilon}^{-\epsilon}=\int du\, \rho(u) u L_{\epsilon}^{\epsilon}(u) K^{-\epsilon}(u) . \quad \text{(A5)}$$

Let us now assume that similarly a field V_n $(n>m)$ acts upon u_m. We have, after (22),

$$\mathbf{Q}_N=(\tilde{\mathscr{C}})^{N-n-1}(\hat{\mathscr{C}}_{n+1})(\tilde{\mathscr{C}})^{n-m-1}(\hat{\mathscr{C}}_{m+1})(\tilde{\mathscr{C}})^{m-1}\mathbf{Q}_1 , \quad \text{(A6)}$$

which for n, m, N, and $N-n$ tending to infinity gives

$$Z_N(V_m,V_n)=(1+\mu\nu\varphi^{n-m-1})Z_N(0,0) , \quad \text{(A7)}$$

where

$$\mu=\beta V_m \frac{f_{-1}^{+1}}{F_{+1}^{+1}}, \quad \nu=\beta V_n \frac{f_{+1}^{-1}}{F_{+1}^{+1}}, \quad \varphi=\frac{F_{-1}^{-1}}{F_{+1}^{+1}} . \quad \text{(A8)}$$

We thus obtain immediately the correlation

$$\langle u_m u_n \rangle = \beta^{-2}\left[\frac{\partial^2}{\partial V_m \partial V_n} Z_N(V_m,V_n)\right]_{V_m=V_n=0} , \quad \text{(A9)}$$

that is,

$$\langle u_m u_n \rangle = \frac{(F_{+1}^{+1})^2}{f_{-1}^{+1} f_{+1}^{-1}}\varphi^{n-m-1} . \quad \text{(A10)}$$

If we define the correlation length ξ by

$$\xi^2=\frac{\sum_{p=-\infty}^{+\infty} p^2\langle u_n u_{n+p}\rangle}{\sum_{p=-\infty}^{+\infty} \langle u_n u_{n+p}\rangle} , \quad \text{(A11)}$$

we obtain, in the low-temperature limit,

$$\xi=\sqrt{2}(1-\varphi)^{-1} . \quad \text{(A12)}$$

APPENDIX B

We start again with a ferrimagnetic chain showing translational symmetry and no external field. Now, we introduce $\mathbf{B}$ acting on the v component $(v=x,y)$ of $\mathbf{S}_m$ only. This merely changes $\phi_m(u_{m-1},u_m)$ from $\tilde{\phi}_m(u_{m-1},u_m)$ to

$$\hat{\phi}_m(u_{m-1},u_m)=\tilde{\phi}_m(u_{m-1},u_m)\psi(u_m) , \quad \text{(B1)}$$

where

$$\psi(u_m)=\int dv \rho'(v,u_m)\exp(-\beta G\mu_B S v B) . \quad \text{(B2)}$$

In these expressions $\rho'(v,u)$ is the distribution of the v component of $\mathbf{S}_m/S$ when S_m^z/S is fixed at u. For small enough applied field, this reduces to

$$\psi(u_m)=1+(\beta G\mu_B SB)^2\overline{v^2} \quad \text{(B3)}$$

where $\overline{v^2}$ is the mean value of v^2 with the distribution $\rho'(v,u_m)$. This vanishes for $d=1$; for $d=2$, this vanishes too if $\mathbf{B}$ is along $\hat{\mathbf{y}}$, and is simply $1-u_m^2$ if $\mathbf{B}$ is along $\hat{\mathbf{x}}$. For $d=3$, this is $(1-u_m^2)/2$ whatever the direction of $\mathbf{B}$ normal to $\hat{\mathbf{z}}$. We thus have

$$\psi(u_m)=1+\frac{k_v^{(d)}}{4}(\beta G\mu_B SB)^2(1-u_m^2) , \quad \text{(B4)}$$

with $k_v^{(d)}=0,1,2$ depending on the situation under consideration. Since this is an even function of u_m, it only affects L_{m+1}^{+1} and L_{m-1}^{+1}. As a result, the largest eigenvalue of $(\mathscr{C}_{m+1})$ and finally the partition function as a whole are merely multiplied by the factor

$$X=\frac{\int du\, \rho(u)L_{+1}^{+1}(u)K^{+1}(u)\psi(u)}{\int du\, \rho(u)L_{+1}^{+1}(u)K^{+1}(u)} . \quad \text{(B5)}$$

Thus, we get simply

$$\chi_{v,c}^{(d)}=-\frac{1}{\beta r^2}\left[\frac{\partial^2 \ln X}{\partial B^2}\right]_{B=0} , \quad \text{(B6)}$$

that is,

$$\chi_{v,c}^{(d)}=\beta k_v^{(d)}(G\mu_B S)^2\frac{\int du\, \rho(u)(1-u^2)L_{+1}^{+1}(u)K^{+1}(u)}{\int du\, \rho(u)L_{+1}^{+1}(u)K^{+1}(u)} . \quad \text{(B7)}$$

These quantities have analytical expressions in terms of Bessel functions, as given in Table II.

APPENDIX C

Let now the field $\mathbf{B}$ act on the v component of $\mathbf{s}_m$ $(v=x,y)$. The function $\phi_m(u_{m-1},u_m)$ is now changed from $\tilde{\phi}_m(u_{m-1},u_m)$ to

$$\hat{\phi}_m(u_{m-1},u_m)=\mathrm{Tr}[\exp(-\beta(H_m-g\mu_B B s_m^v))] , \quad \text{(C1)}$$

where the trace is over the states of $\mathbf{s}_m$. Computing it on the basis of the eigenstates of $\mathbf{s}_m$ submitted to the effective (exchange plus external) field we get

$$\hat{\phi}_m(u_{m-1},u_m)=\phi(u_{m-1},u_m)+\frac{\beta}{2}\frac{(g\mu_B)^2}{|J|S}\frac{\sinh[a/2(u_{m-1}+u_m)]}{u_{m-1}+u_m} . \tag{C2}$$

Clearly, this expression does not fit the standard form (9). However, this is not a major difficulty since only one site is actually affected. Developing the extra part of $\hat{\phi}_m$ into a power series of $(u_{m-1}+u_m)$, we get terms which all fit the standard form and enter the general frameworks described in Appendices A and B. Moreover, due to the parity of the development the various terms are either even, or odd, with respect to u_{m-1} and u_m, simultaneously. As a result, only the diagonal terms of $(\mathscr{C}_{m-1})$ are changed, whereas the off-diagonal one remains equal to zero. Since only the even contributions both affect the largest eigenvalue and contribute to the integral of interest,

$$J_4=\int du\,dv\rho(u)\rho(v)\left[\frac{a}{2}(u+v)\right]^{-1}\cosh\left[\frac{au}{2}\right]\times\cosh\left[\frac{av}{2}\right]\sinh\left[\frac{a}{2}(u+v)\right] , \tag{C3}$$

it is readily shown that the contribution of the extra part of $\hat{\phi}$ to the partition function results in the simple factor

$$Y=1+\tfrac{1}{4}\beta^2(g\mu_B)^2J_4J_1^{-2} \tag{C4}$$

(J_1 is defined in the text). As a result, we get immediately,

$$\chi_{x,Q}^{(d)}=\chi_{y,Q}^{(d)}=\beta\left[\frac{g\mu_B}{2}\right]^2J_4J_1^{-2} . \tag{C5}$$

Unfortunately, J_4 may not be given a simple expression in terms of S, J, T, and d. In Appendix D we derive practical series expansions for this quantity.

APPENDIX D

In order to evaluate the quantity J_4 we introduce the function

$$K(u,v)=\int_{-1}^{+1}\rho(t)\cosh[(u+v)t]+\cosh[(u-v)t]dt . \tag{D1}$$

Then we have

$$J_4=+\frac{1}{2a}\int_0^{a/2}\left[K\left[\frac{a}{2},v\right]\right]^2dv . \tag{D2}$$

The integral $K(u,v)$ is easily calculated using the expressions for $\rho(u)$ given in Sec. II. We obtain

$$K(u,v)=a^{\delta}\Lambda_d\left[\frac{I_\delta(u+v)}{(u+v)^\delta}+\frac{I_\delta(u-v)}{(u-v)^\delta}\right] , \tag{D3}$$

with δ and Λ_d given in Table I. Then J_4 may be written

$$J_4=\frac{1}{2a}a^{\delta}\Lambda_d(S_1+2S_2) , \tag{D4}$$

where

$$S_1=\int_0^{a/2}\left[\frac{I_\delta^2[(a/2)+v]}{[(a/2)+v]^{2\delta}}+\frac{I_\delta^2[(a/2)-v]}{[(a/2)-v]^{2\delta}}\right]dv , \tag{D5}$$

$$S_2=\int_0^{a/2}\left[\frac{I_\delta[(a/2)+v]I_\delta[(a/2)-v]}{[(a/2)+v]^{\delta}[(a/2)-v]^{\delta}}\right]dv . \tag{D6}$$

Using the series expansion[18]

$$I_\delta^2(x)=\sum_{n=0}^{\infty}\frac{(2n+2\delta)!(x/2)^{2(n+\delta)}}{(n+\delta)!n!(n+2\delta)!} \tag{D7}$$

and the relation

$$(2y)!=\pi^{-1/2}2^{2y}y!(y-\tfrac{1}{2})! , \tag{D8}$$

we get immediately

$$S_1=\pi^{-1/2}\sum_{n=0}^{\infty}\frac{(n+\delta-\frac{1}{2})!a^{2n+1}}{(2n+1)n!(n+\delta)!(n+2\delta)!} . \tag{D9}$$

The integral S_2 may be evaluated using the series expansion of the Bessel functions:

$$I_\delta(x)=\sum_{n=0}^{\infty}\frac{(x/2)^{\delta+2n}}{n!(n+\delta)!} . \tag{D10}$$

We get

$$S_2=\sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\frac{2^{-2(m+n+\delta)}S_{nm}}{n!(n+\delta)!m!(m+\delta)!} , \tag{D11}$$

with

$$S_{nm}=\tfrac{1}{2}\int_0^{a/2}\left[\left[\frac{a}{2}+v\right]^{2m}\left[\frac{a}{2}-v\right]^{2n}+\left[\frac{a}{2}+v\right]^{2n}\left[\frac{a}{2}-v\right]^{2m}\right]dv . \tag{D12}$$

Now, from the properties of the beta function we can deduce[18]

$$\int_0^1 t^{k-1}(1-t)^{l-1}dt=\frac{(k-1)!(l-1)!}{(k+l-1)!} \tag{D13}$$

and

$$S_{nm}=\tfrac{1}{2}a^{2m+2n+1}\frac{(2m)!(2n)!}{(2m+2n+1)!} . \tag{D14}$$

Finally, S_2 is expressed as

$$S_2=\pi^{-1}2^{-1-2\delta}\sum_{N=0}^{\infty}\frac{a^{2N+1}}{(2N+1)!}R_{\delta,N},\quad N=n+m , \tag{D15}$$

where $R_{\delta,N}$ is the result of the finite summation

$$R_{\delta,N}=\sum_{n=0}^{N}\frac{(n-\frac{1}{2})!(N-n-\frac{1}{2})!}{(n+\delta)!(N-n+\delta)!} . \tag{D16}$$

Considering the δ values of interest, we have

$$R_{-1/2,n}=n+1\ , \tag{D17}$$

$$R_{0,n}=\pi\ , \tag{D18}$$

$$R_{1/2,n}=\frac{4}{n+1}\sum_{l=0}^{n}(2l+1)^{-1}\ . \tag{D19}$$

Finally, for $d=1$, we have simply

$$J_4=(8a)^{-1}[\sinh(2a)+2\sinh a+2a\cosh a+2a]\ , \tag{D20}$$

and in the other cases,

$$J_4=\frac{1}{2a}a^{\delta}\Lambda_d\sum_{n=0}^{\infty}\frac{a^{2n+1}}{2n+1}\left[\pi^{-1/2}\frac{(n+\delta-\frac{1}{2})!}{n!(n+\delta)!(n+2\delta)!}+\frac{R_{\delta,n}}{2^{\delta}\pi(2n)!}\right]\ , \tag{D21}$$

with $R_{\delta,n}$ given by (D18) and (D19).

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