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Ferrimagnetic Heisenberg chains [$\frac{1}{2}$-S] ($S=1$ to $\frac{5}{2}$): Thermal and magnetic properties

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Energy spectra and thermodynamic quantities (partition function, entropy, specific heat, susceptibility) of ferrimagnetic Heisenberg chains, made up of two spin sublattices [$\frac{1}{2}$-S], are discussed from the exact results computed for finite rings. Extrapolation of the theoretical data versus the length of finite rings N is discussed for S ranging from 1 to $\frac{5}{2}$ and compared with previous reported findings for the [$\frac{1}{2}$-∞] spin chain. In the low-temperature region, similarities between the ferrimagnetic chains [$\frac{1}{2}$-S] and the regular ferromagnetic ones (with spin S-$\frac{1}{2}$) are used for the determination of the limiting curves.

I. INTRODUCTION

In spite of extensive literature, the one-dimensional exchange-coupled systems always offer a great challenge to physicists and chemists for describing phenomena that cannot be explained in higher dimension.[1–4] Several physical situations were inspected and solved rigorously, although some of them appear, to a certain extent, to be of purely academic interest. Thus, closed expressions of the thermodynamic quantities were derived when the interactions between nearest neighbors spread out in a one-dimensional (1D) Ising network[5–7] or in an XY anisotropic one.[8] Likewise, the Heisenberg-chain problem was shown to be soluble analytically[9] only in the classical limit, $S\rightarrow\infty$. Unfortunately, an exact treatment is not available when an isotropic interaction is assumed in the quantum chain. The limiting behavior may then be estimated from the numerical results of finite closed chains (rings) extrapoled to the thermodynamic limit $N\rightarrow\infty$. Such a method was initiated by Bonner and Fisher[10] for the $S=\frac{1}{2}$ regular chain and subsequently extended by Weng[11] and Blöte[12] to arbitrary spin quantum numbers. The same computational procedure was used by Duffy and Barr[13] for solving the alternately spaced $S=\frac{1}{2}$ chain, involving two distinct exchange parameters between nearest neighbors.

Although several works have focused on the behavior of exotic chains, and despite the large collection of 1D materials, ferrimagnetic chains made up of two unequal spin sublattices have not been extensively investigated. A first analysis of the correlation functions and specific heat was suggested by Dembinski and Wydro[14] in the particular case of a quantum-classical Heisenberg chain. Further, Blöte[15] determined the expression of the susceptibility by neglecting the contribution of the quantum sublattice with respect to the classical one, making this treatment unadaptable for real systems.

The recent discovery of ordered bimetallic chain compounds has given rise to a significant development of this challenging problem.[16–24] For the first time, we have focused on the [$\frac{1}{2}$-1] Heisenberg spin chain,[25] the solutions of which were computed for finite rings of increasing size N. The calculations were performed up to ten spins for distinct values of the g_a and g_b Landé factors, and then extrapoled to the thermodynamic limit ($N\rightarrow\infty$). Independently, Verdaguer *et al.*[26] have solved this problem up to eight spins by neglecting Zeeman contributions due to alternating Landé factors ($g_a\neq g_b$); further Seiden *et al.*[27] have solved numerically the magnetic susceptibility of the [$\frac{1}{2}$-S] chain when S is a classical spin, and more recently Georges *et al.*[24] have derived analytical solutions for the J-alternating [s-S] system, s being an arbitrary quantum spin. Lastly, we have proposed in the classical limit (both spin sublattices are classical vectors which only differ through Landé factors) an analytical expression of the susceptibility which was shown to be convenient for large spin systems.[28] Further, in the case of anisotropic exchange (Ising-type coupling), exact solutions have been derived for a large number of spin configurations, including local anisotropies and alternating exchange.[29–31] The most striking feature predicted by this model is the occurrence of a compensation temperature, similar to that of ferrimagnetic garnets, although long-range ordering can only be observed at absolute zero.[30]

The study reported here deals with thermal and magnetic properties of [$\frac{1}{2}$-S] Heisenberg chains by assuming an antiferromagnetic exchange coupling between nearest neighbors, only. Numerical results will be discussed for S ranging from 1 to $\frac{5}{2}$ and compared to the classical limit $S\rightarrow\infty$.

II. EIGENVALUE PROBLEM

We consider a Heisenberg quantum chain made up of two spin sublattices, namely S_a and S_b. Let S_i ($i=1,2,\ldots,2N$) be the current spin vector with values S_a or S_b depending on whether the site parity is odd or even. Assuming a finite closed chain of $2N$ spins, the exchange Hamiltonian is expressed as

$$H=-J\sum_i S_{2i}(S_{2i-1}+S_{2i+1}) ,$$

where a negative J value refers to an antiferromagnetic coupling and cyclic boundary conditions impose $S_{2N+1}=S_1$.

For finite strings of N pairs (S_a-S_b), the eigenvalue problem consists of solving a $(2S_a+1)^N\times(2S_b+1)^N$ energy matrix. As shown in Ref. 25, significant reduction of the computational work is obtained by fully taking into account the geometrical and spin space symmetries of the $2N$-site closed chain. Then, eigenfunctions of H transform according to the irreducible representations of the point group D_N, instead of D_2 when assuming linear segments.

Considering the translation operator T, which transforms the site i into $i+2$, and the mirror operator I, which transforms i into $2(N+1)-i$, we can define symmetrized Bloch states which result in a much more tractable eigenvalue problem. These operations drastically reduce the size of the largest block matrix to be diagonalized by a factor depending on the spin multiplicities (Table I). For the $[\frac{1}{2}\text{-}1]_5$ closed chain, for instance, the size of the largest block to be solved is 76×76 instead of 7776×7776, when no symmetry is involved.

TABLE I. Matrix dimension for the [$\frac{1}{2}$-S] chains.

System	N	Largest block	Reduction factor
[$\frac{1}{2}$-1]	5	76	100
[$\frac{1}{2}$-$\frac{3}{2}$]	4	22	190
[$\frac{1}{2}$-2]	4	65	150
[$\frac{1}{2}$-$\frac{5}{2}$]	3	13	130

III. DISCUSSION OF RESULTS

A. Energy spectrum

The procedure for estimating thermal and magnetic behaviors in the thermodynamic limit ($N\to\infty$) from those of finite closed chains was discussed in the case of regular antiferromagnetic chains.[10,12,32] It needs finding correlations between the calculated results for finite closed chains, depending on the size N.

Thus, the ground-state energies of finite rings (Table II), normalized to the ($\frac{1}{2}$-S) pair, are assumed to be related to those of the infinite system by the relationship

$$E_N(0)=E_\infty(0)+a/N^p ,$$

where p is real and positive. Note, in this case, that the determination of $E_\infty(0)$ is more difficult than for the regular antiferromagnetic chain,[32] since $E_\infty(0)$ is approached monotonically by lower values, only. Anyway, it is rather well defined for the [$\frac{1}{2}$-1] system due to the rapid convergence of the levels when N increases. Further, it may be noticed that the exponent p is not an integer as suggested for regular antiferromagnetic chains.

When dealing with $[\frac{1}{2}\text{-}S]_N$ systems with larger S, we may expect that the reduction of the maximum chain length results in a poor estimate of $E_\infty(0)$; for example, the ground-state energy of the infinite $[\frac{1}{2}\text{-}\frac{5}{2}]_N$ system is determined from the limited sequence $N=1$ to 3. The rigorous treatment for $S\to\infty$ (quantum-classical chain) shows, in fact, that a two-spin unit enables us to describe the behavior of the infinite system; a smoother dependence of the properties with N is thus expected when S becomes large enough.

The comparison of the ground-state energies is reported for a sequence of S values as $E_\infty(0)/NJ(S\text{-}\frac{1}{2})$ versus $(S\text{-}\frac{1}{2})^{-1}$ (Fig. 1). The limiting value ($S\to\infty$) corresponds to the exact solutions of the quantum-classical chain solved by Dembinski and Wydro:[14]

$$E_\infty(0)/NJ=-(1/x)\ln[2/x^2(x\sinh-\cosh x+1)]$$

with $x=|J|/kT$, which tends towards unity as $T\to0$ K. Clearly, this value is very well fitted (the accuracy is better than 2%) by linear extrapolation of $E_\infty(0)$ versus $(S\text{-}\frac{1}{2})^{-1}$, justifying the above remarks when S increases.

The determination of the partition function is based on the knowledge of the complete set of eigenvalues for each $[\frac{1}{2}\text{-}S]_N$ ring. As shown in Fig. 2 for $[\frac{1}{2}\text{-}1]_N$, the number of levels increases drastically with N, giving a quasicontinuous spectrum for $N=5$ (not reported on the graph). Closer examination shows that the density of levels becomes swiftly substantial in the intermediate region of the spectrum, while these are quite spaced in the vicinity of the ground state. An energy continuum being expected as $N\to\infty$, it is clear that the extrapolation procedure of the thermodynamic functions may lead to severe devi-

TABLE II. Ground-state energy of [$\frac{1}{2}$-S] N rings.

$E_N(0)/NJ$ system	$N=1$	$N=2$	$N=3$	$N=4$	$N=5$	$N=\infty$
[$\frac{1}{2}$-1]	2.0000	1.5000	1.4626	1.4560	1.4546	1.452
[$\frac{1}{2}$-$\frac{3}{2}$]	2.5000	2.0000	1.9716	1.9680		1.965
[$\frac{1}{2}$-2]	3.0000	2.5000	2.4760	2.4740		2.472
[$\frac{1}{2}$-$\frac{5}{2}$]	3.5000	3.0000	2.9800			2.975

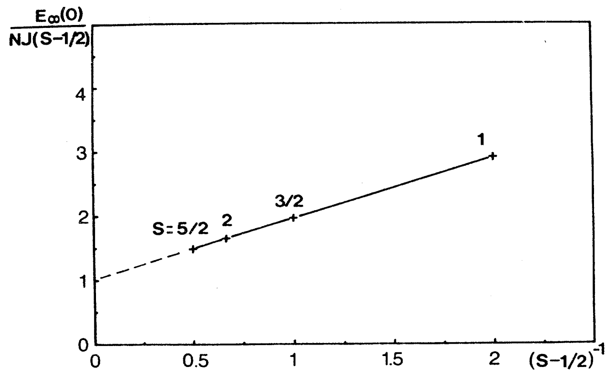

FIG. 1. Ground-state energies of $[\frac{1}{2}\text{-}S]$ ferrimagnetic chains.

ations from the real behavior when approaching absolute zero. Further, it may be noticed that the multiplicity of the low-lying levels (corresponding to the length of the straight lines on the graph) decreases continuously from $N+1$ to 1 or 2, according to N parity. Thus, the very low-temperature diagram closely resembles that of a N spin-$\frac{1}{2}$ chain with a ferromagnetic exchange coupling between nearest neighbors. Such an analogy, already emphasized in a previous paper,[25] was used for very low-temperature extrapolations. Obviously, the comparison with the ferromagnetic $S=\frac{1}{2}$ chain becomes totally irrelevant at higher temperatures.

B. Thermal properties

Let us examine the thermal behavior of $[\frac{1}{2}\text{-}1]_N$ rings with N running from 1 to 5. A plot of the internal energy $(U/N|J|)$ versus $kT/|J|$ is given in Fig. 3. We note that for increasing N: (i) the limiting curve is approached monotonically by lower values. (ii) the difference between isothermal energies decreases swiftly with increasing ring sizes. As a result, the limiting energy appears to be reliable in all the temperature ranges with an error lower than 0.5%. In the limit of low $kT/|J|$ values, it may be verified that the power law,

$$U_\infty(T)=U_\infty(0)+AN|J|/x^{3/2}$$

which holds for the $S=\frac{1}{2}$ ferromagnetic chain, is a good approximation. The curves giving the thermal variation of the entropy (S/Nk) are displayed in Fig. 4. Due to the crossing of successive curves in the range $kT/|J|=0.2-0.4$, the determination of the limiting curve appears to be more complicated. At absolute zero, the entropy, which only depends on the ground-state multiplicity, falls to zero for $N\rightarrow\infty$ as $S/Nk=(1/N)\ln(N+1)$.

According to the spin-wave theory, and the computations performed for limited ferromagnetic chains, one may predict the low-temperature behavior to be accurately described by the power law

$$S_\infty(T)=ANk/x^{1/2}\ ,$$

where the power $\frac{1}{2}$ is obviously directly related to that found above for the internal energy. For the $S=\frac{1}{2}$ ferromagnetic chain, Bonner and Fisher found $A=0.85$ while spin-wave theory predicts an amplitude larger by a factor about 1.3 (Ref. 10). An attempt to make a description of the low-temperature behavior from spin-wave theory (dashed curve in Fig. 4) agrees well with the expected trend from limited chain findings. From these results, it is now possible to discuss the specific-heat curves

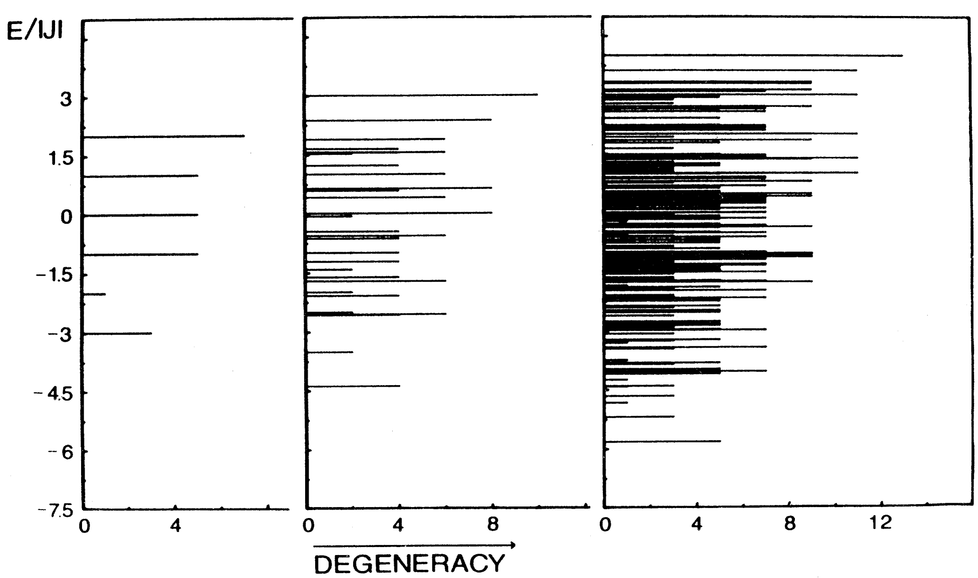

FIG. 2. Energy levels of $[\frac{1}{2}\text{-}1]_N$ rings for N running from 2 to 4.

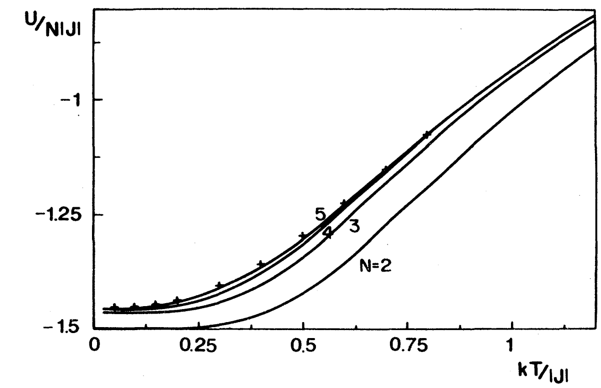

FIG. 3. Internal energy (per spin pair) of $[\frac{1}{2}\text{-}1]$. The limiting curve is displayed as (+).

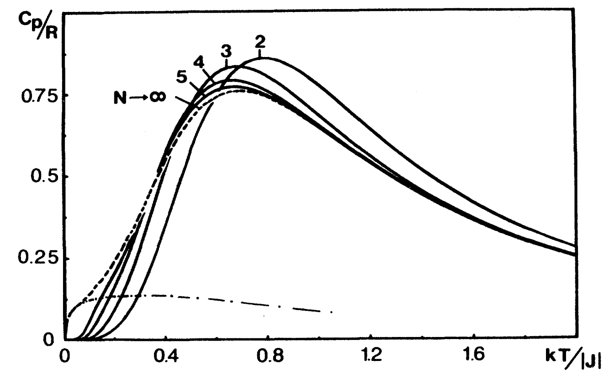

FIG. 5. Specific heat (per spin pair) of $[\frac{1}{2}\text{-}1]_N$ rings. The limiting curve ($N\rightarrow\infty$) is displayed as the dashed line. The $S=\frac{1}{2}$ ferromagnetic chain is reported in dotted-dashed line.

of ferrimagnetic Heisenberg chains. We have plotted in Fig. 5 the specific-heat results for $[\frac{1}{2}\text{-}1]_N$ closed chains.

Unlike the antiferromagnetic chain, the curves for finite N do not bracket the limiting curve, making the extrapolation procedure a more difficult task, at least at low temperature. It can be emphasized that successive curves cross for $kT/|J|$ ranging between 0.3 and 0.6 and show, for $N=5$, an unexpected inflection point at low temperature. Owing to these remarks, it is clear that any extrapolation procedure used at high temperature becomes irrelevant in the crossing region and below. In fact, remembering the analogy with the $S=\frac{1}{2}$ ferromagnetic chain, one can estimate the limiting curve in the very low-temperature region ($kT/|J|<0.2$) from Bonner and Fisher results[10] (dashed-dotted line). It then appears that the specific heat varies as $T^{1/2}$ for $T\rightarrow 0$ K, while we found for finite closed chains an exponential variation. This apparent disagreement results, in fact, from the occurrence of large gaps between low-lying levels in the case of finite chains (see Fig. 2), while in the thermodynamic limit these levels close up to give a continuum, with a high-spin multiplicity. This induces a remarkable phenomenon which is the occurrence of a second anomaly at low temperature for the infinite $[\frac{1}{2}\text{-}1]$ chain. Further, we note that the low-temperature anomaly is significantly enhanced when S increases, as can be observed in Fig. 6, for $[\frac{1}{2}\text{-}2]_N$. Clearly, it is closely related to the spin multiplicity of the basic unit $[\frac{1}{2}\text{-}S]$, so that a drastic evolution of the specific heat might be inferred for the quantum-classical spin chain as $T\rightarrow 0$ K. We have plotted in Fig. 7 the numerical results for $[\frac{1}{2}\text{-}S]$ quantum closed chains together with those of the quantum-classical one. Only the curves corresponding to finite rings are drawn for $S>1$. Due to required computing times, the calculations were only performed up to $N=4$ for $S=\frac{3}{2}$ and 2, and 3 for $S=\frac{5}{2}$, making the extrapolation a more difficult task. Nevertheless, the height and position of the Schottky-type anomaly should by closely approximated. These results are summarized in Table III.

For the quantum-classical chain, the expression of the specific heat is given by[14]

$$C_p/Nk=2+x^2(-x^2-\cosh^2x+\cosh x+x\sinh x)/(x\sinh x-\cosh x+1)^2\ .$$

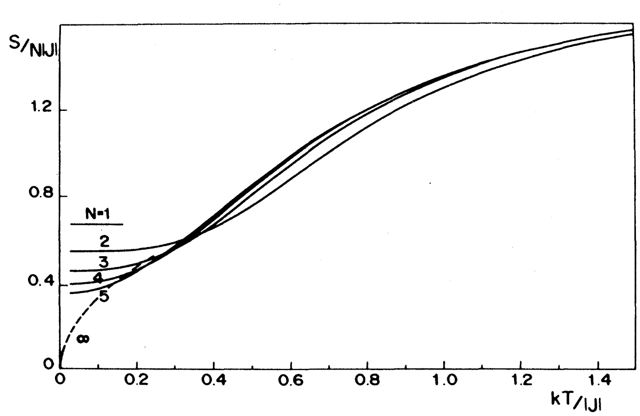

FIG. 4. Magnetic entropy (per spin pair) of $[\frac{1}{2}\text{-}1]_N$ rings. The limiting curve calculated from the spin-wave theory is given in dashed line.

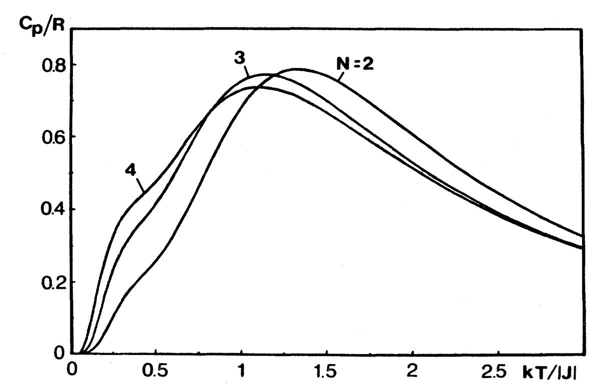

FIG. 6. Specific heat (per spin pair) of $[\frac{1}{2}\text{-}2]_N$ rings.

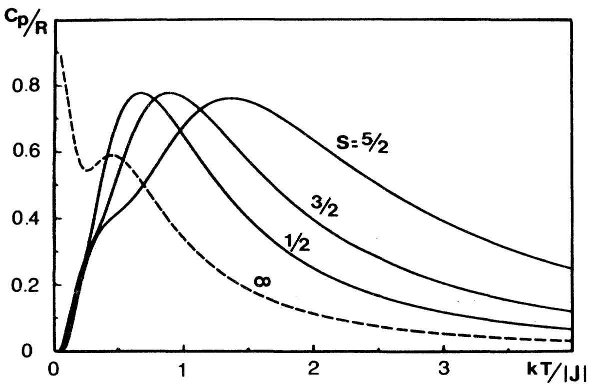

FIG. 7. Specific heat of $[\frac{1}{2}\text{-}S]_N$ rings showing the enhancement of the low-temperature anomaly when S increases. The classical result is displayed in dashed line.

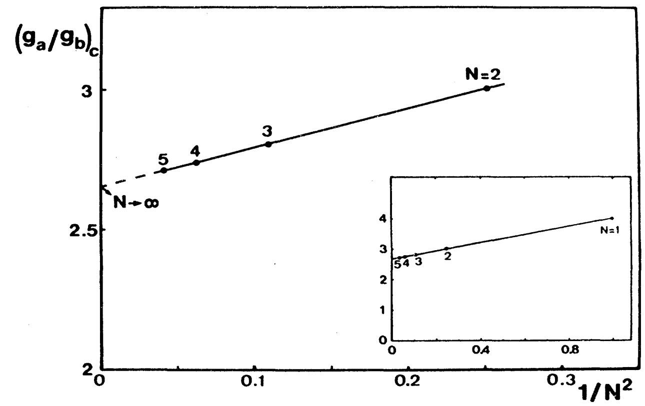

FIG. 8. Critical g_a/g_b values of $[\frac{1}{2}\text{-}1]_N$ rings giving zero magnetic moment in the ground state.

We note that a Schottky-type anomaly is still present in the same range of $kT/|J|$, but the low-temperature variation drastically differs. Thus, C_p shows a nonzero value at $T=0$ K, originated by the classical spin sublattice, which obviously prevents any comparison with real systems.

C. Magnetic properties

The magnetic behavior of finite rings $[\frac{1}{2}\text{-}1]_N$ has already been discussed in Ref. 25. It was emphasized that a nonzero magnetic moment is stabilized in the ground state, except when the ratio between Landé factors (r_g) approaches a critical value, $(r_g)_c$. For this value, the systems behave like antiferromagnetic species in the very low-temperature limit. For the infinite chain, the critical value was approximated from the plot of $(r_g)_c$ versus $1/N^2$ (Fig. 8), giving $(r_g)_c=2.66$. From the same study for the other $[\frac{1}{2}\text{-}S]$ Heisenberg chains, the spin dependence of the critical r_g value was shown to be given by the relationship $(r_g)_c=\frac{4}{3}(S+1)$. This result can be compared to that obtained when the exchange is Ising like.[30] In that case, we show that the compensation of both sublattices arises for $g_a/g_b=2S$. The origin of this difference lies within the larger degrees of freedom of Heisenberg spins with respect to Ising ones.

TABLE III. Thermal and magnetic properties of $[\frac{1}{2}\text{-}S]$ Heisenberg ferrimagnetic chains.

	Specific heat		Magnetic susceptibility[a]	
System	$Cp_{\max}/R$	$kT_{\max}/\|J\|$	$X_{n,\min}$	$kT_{\min}/\|J\|$
$[\frac{1}{2}\text{-}1]$	0.377	0.705	1.507	0.577
$[\frac{1}{2}\text{-}\frac{3}{2}]$	0.356	0.96	2.221	1.190
$[\frac{1}{2}\text{-}2]$	0.351	1.2	2.609	2.1
$[\frac{1}{2}\text{-}5]$	0.35	1.4	2.80	2.9
$[\frac{1}{2}\text{-}\infty]$[b]			2.83	2.98

[a]Values assuming $g_a=g_b$ $(r_g=1)$.
[b]Classical spin scaled to $S=\frac{5}{2}$.

Spin waves are known to provide a convenient description of low-lying magnetic excitations and neighboring spin correlations at low temperature, but they fail to depict accurately all the features of the ground state. The compensation phenomenon deals with both long-range and short-range effects, and it was interesting to examine to which extent this theory can account for it.

We have derived the critical Landé factor ratio from the ferrimagnetic spin-wave spectrum,[33]

$$g_a/g_b=(S_b+\tfrac{1}{2}-X)/(S_a+\tfrac{1}{2}-X)$$

with, for 1D ferrimagnets:

$$X=\frac{1}{2\pi}\int_0^{\pi}(S_a+S_b)/(S_a^2+S_b^2-2S_aS_b\cos x\,dx)^{1/2}\ .$$

In the case of $[\frac{1}{2}\text{-}1]$ chain, we get $g_a/g_b=3.56$ which is significantly larger than the genuine value 2. This shows that the origin of this effect lies probably in the ground-state spin reduction which is the larger, the smaller the spin. The fact that spin-wave theory ignores the interactions between the various modes is doubtless at the origin of the discrepancy between this value and our extrapolated estimate.

The temperature dependence of the magnetic behavior is now discussed in terms of reduced X_nT product

$$X_nT=10XT/[(N_a\mu^2/k)g_b^2\tfrac{3}{4}r_g^2+S(S+1)]\ ,$$

which uniformly takes the value $\frac{10}{3}$ in the high-temperature limit. In this expression, X represents the magnetic susceptibility per spin pair.

Figures 9 and 10 display the behavior of $[\frac{1}{2}\text{-}1]_N$ closed chains, for $r_g=1$ and 2. In the former case, XT decreases as T is raised (except for $N=1$), then presents a rounded minimum near $kT/|J|=0.5$, which is the signature of a ferrimagnetic chain. The extrapolated curve $(N\rightarrow\infty)$, drawn as a dashed line, has been estimated from well-known procedures.[32] It is worth noticing that around the minimum and at a higher temperature, the variation of the limiting curve closely coincides with that of the $[\frac{1}{2}\text{-}1]_5$ ring. Below this minimum, the observed divergence

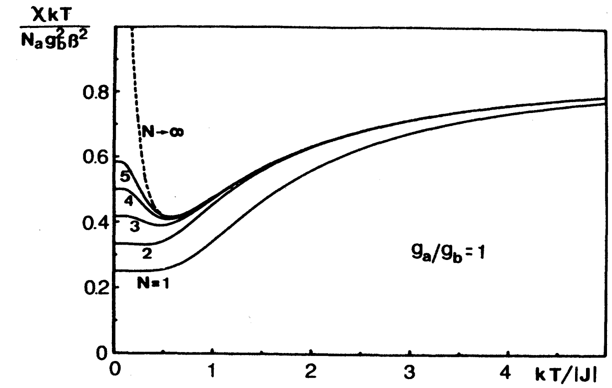

FIG. 9. Magnetic behavior (per spin pair) of $[\frac{1}{2}\text{-}1]_N$ when $g_a/g_b=1$. The limiting behavior ($N\rightarrow\infty$) is displayed as the dashed line.

obeys, at least down to $kT/|J|=0.2$, the power law $T^{-0.8}$ in good agreement with the behavior of the $S=\frac{1}{2}$ ferromagnetic chain. We can assume that this variation holds at lower temperature, in the range where any extrapolation becomes questionable.

Figure 10 shows the variation of the magnetic susceptibility when approaching the critical r_g value. X_n exhibits a rounded maximum at $kT/|J|=0.8$, followed by a minimum and a divergence upon cooling down. Similarly to the previous case, it can be noticed that the $N=5$ curve closely describes the infinite system down to $kT/|J|=0.5$. For subsequent comparisons with any experiment, we report in Table IV polynomials obtained by fitting the limiting curves for $r_g=1$ and 2.

Following the same procedure, we have computed the susceptibility of ferrimagnetic chains [$\frac{1}{2}$-S] for $S>1$.

The fact that the calculations for $S=\frac{5}{2}$ were limited to $N=3$ has prevented us to do any satisfying extrapolation at low temperature. Anyway, it is worth noticing that data for large S converge more rapidly, when N increases, than those for small S. Thus, the convergence is rather rapid above the XT minimum suggesting that, above this temperature, the curve for $N=3$ describes the limiting

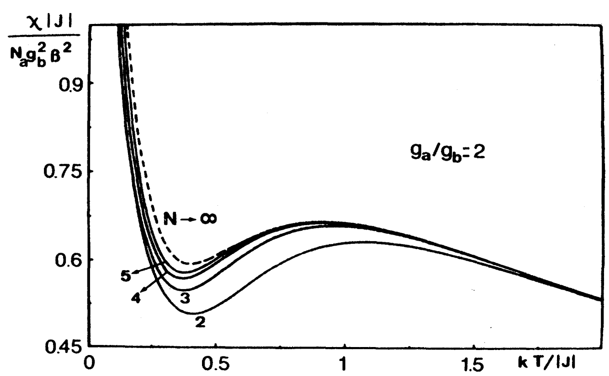

FIG. 10. Magnetic behavior (per spin pair) of $[\frac{1}{2}\text{-}1]_N$ when $g_a/g_b=2$. The limiting behavior ($N\rightarrow\infty$) is displayed as the dashed line.

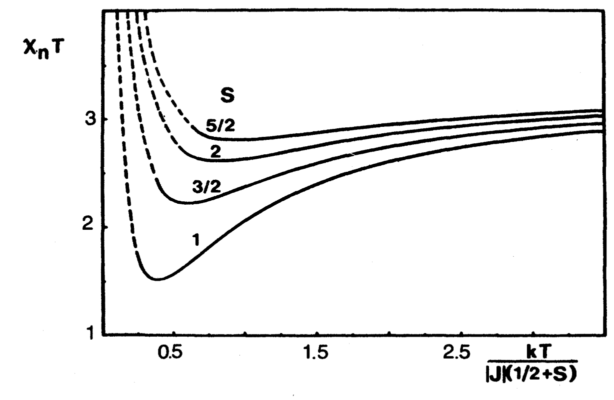

FIG. 11. Magnetic behavior (per spin pair) of [$\frac{1}{2}$-S] Heisenberg ferrimagnetic chains.

behavior in a satisfying manner.

The magnetic behavior of the [$\frac{1}{2}$-S] ferrimagnetic chains is plotted in Fig. 11. All the curves show a typical minimum of XT, which is less pronounced as S is increased; Table III gives the coordinates of minima. Further, at decreasing temperature, the behavior looks like that of regular ferromagnetic chains (dashed lines). Finally, it is worthwhile to notice that the behavior of the [$\frac{1}{2}$-$\frac{5}{2}$] chain rigorously coincides that of the [$\frac{1}{2}$-∞] system[27] in a large region of temperature ($kT/|J|>0.5$); thus, both models predict, within the experimental error, the same coordinates for the XT minimum (see Table III). This justifies the use of the classical model to describe the magnetic behavior of 1D ferrimagnets containing at least one large spin sublattice.

To conclude, it may be emphasized that several bimetallic chain compounds exhibiting such a specific behavior were investigated in the past few years.[16–24] Note also that very close magnetic behaviors were recently reported in the case of homometallic chains,[20,21] due either to noncompensating Landé factors in regular chains or to

TABLE IV. Polynomials giving the behavior of [$\frac{1}{2}$-1] Heisenberg chains for regular ($g_a=g_b$) and alternating ($g_a=2g_b$) Landé factors.

$r_g=g_a/g_b$	$\dfrac{XT^{a}}{(N_a\mu^2/k)g_b^2}$
1	$\dfrac{Ax^3+Bx^2+Cx+D}{Ex^2+Fx+G}$ $A=-0.034\,146\,801$, $B=2.816\,930\,6411$, $C=-7.231\,001\,3697$, $D=11$, $E=1.296\,632\,74$, $F=0.697\,190\,135\,95$, $G=12$
2	$\dfrac{Ax^2+Bx+C}{Dx^3+Ex^2+Fx+G}+Hx$ $A=2.944\,723\,391$, $B=-8.643\,216\,582$ $C=20$, $D=2.207\,977\,566$, $E=2.210\,070\,570$, $F=5.150\,935\,691$ $G=12$, $H=0.002\,323\,25$

[a] $x=|J|/kT$.

topology effects.[34] In the former, it is clear that alternating g factors in a S-spin chain must induce different magnetic moments on successive sites. The ground state is $S=0$ for an antiferromagnetic coupling, so that the low-temperature behavior uniquely results from mixing in between different spin states, namely second-order Zeeman contributions. On the other hand, particular arrangements of identical spins on a 1D network, like intertwinning double chains, can stabilize a ground state in which spin sublattices do not compensate each other.[35] The ferrimagnetism is then purely of topological origin. The theoretical study of such systems shows that all the features of ferrimagnetic chains are still present, and that their behavior may, in some cases, be approximated by that of $[\frac{1}{2}\text{-}S]$ chains reported here.

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