# Ferromagnetic Heisenberg chains: A description of the magnetic susceptibility from a noncritical scaling approach

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A noncritical scaling model is reported to describe the behavior of low-dimensional magnetic systems. Unlike the classical phase transition approach, nonsingular solutions are deduced that are worthwhile when correlations exist, but which are not large enough to trigger a long range order at $T\neq 0$. The notion of "universality class" is extended to these systems that stand at or below a lower critical dimensionality, and illustrated for the quantum Heisenberg ferromagnetic chain. In this system, the $T$ dependence of $\chi T$ exhibits a power law with a negative critical exponent, $-1.25S$, and a negative critical temperature.

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## I. INTRODUCTION

Since the 1960s a lot of work has been devoted to phase transitions and critical phenomena that reveal the existence of striking similarities in the behavior of very different physical systems.[1] Most of the experiments show that, superconductors apart,[2] the behavior of the order parameter is very different from that predicted by the Landau theory,[3,4] pointing out the key role played by fluctuations.

Magnetic phase transitions have been extensively investigated because of the large variety of materials showing different spins (Ising, XY, or Heisenberg) and lattice dimensionalities.[5–7] An important feature is that in the vicinity of the ordering temperature, $T_C$, magnetic moments become strongly correlated within domains whose mean size $\xi$ diverges as a power law of $1/(T-T_C)$. Accordingly, the variation of the thermodynamic functions reflects that divergence, being also described by power laws with a critical exponent $\gamma$, which depends on a few relevant parameters, namely the dimension of the system $(d)$, the order parameter $(n)$, and the range of the interaction.[8,9] This induces two key ideas into the critical phenomena theory, namely the concept of scaling invariance and the concept of universality.

We have previously reported a model of "hierarchical superparamagnetism," that generalizes the ideas of scaling, by allowing for nonsingular solutions, together with the singular ones, in the classical "critical scaling" approach.[10–12] Nonsingular solutions, that are usually ruled out, are shown to be worthwhile when correlations exist, but which are not large enough to trigger a long-range magnetic order at a finite $T_C$, because we sit at, or below, a lower critical dimensionality, as evidenced in the Heisenberg spin-1 Haldane chain or the two-dimensional (2D) Heisenberg systems.[13–15]

In the present paper, a noncritical scaling model is applied to the ferromagnetic quantum chain of isotropic spins $S$, which can only be solved numerically for finite units, and we show that the magnetic susceptibility is very well described by a universal power law.

## II. NONCRITICAL SCALING: THE OTHER SOLUTIONS OF THE SCALING MODEL

We consider in what follows a ferromagnetic system made of $N$ individual units of size $\xi_0$, and magnetic moment $\mu=[S(S+1)]^{1/2}$ (normalized to $g^2\mu_B^2/k$). Due to the interaction between nearest neighbors, these moments align when the temperature is low enough, and the correlation length, $\xi(T)$, defines the size of new objects. Their volume is $\xi^d$ in space dimension $d$, and their number $N=N_0(\xi/\xi_0)^{-d}$. Accordingly, the magnetic moment, which increases like the volume of the objects (only true for a ferromagnet), may be written as

$$\chi T \propto \xi(T)^d \tag{1}$$

The "static scaling hypothesis" then assumes that $\xi$ increases when $T$ decreases, and eventually diverges like a power of $(J/T_C-J/T)$. This requires that

$$\xi/\xi_0 = (1-T_C/T)^{-\nu} = (1-T_C/T)^{-\Theta/T_C} \tag{2}$$

with $\Theta=\nu T_C>0$, which may be written as

$$\ln(\xi/\xi_0) = \Theta/T + \Theta T_C/2T^2 + \Theta T_C^2/3T^3 + \cdots . \tag{3}$$

In the corresponding Arrhenius plot of $\xi/\xi_0$, all solutions are monotonous functions of $1/T$ which increase when $T$ decreases, with an initial slope which is precisely $\Theta$.

For $\Theta>0$, the sign of $T_C$ fixes the curvature in such a way that the Arrhenius representation of $\xi/\xi_0=\exp(\Theta/T)$, corresponding to the $T_C=0$ limit, separates solutions of positive curvature, $\xi/\xi_0=(1-T_C/T)^{-\Theta/T_C}$, from those whose curvature is negative, $\xi/\xi_0=(1+T_K/T)^{\Theta/T_K}$ where $T_C=-T_K$. Neither of these solutions is forbidden by the thermodynamics.

The "static scaling assumption" has measurable consequences. In particular, using Eq. (1) and permitting $T_C$ to be positive null or negative, we obtain

$$\chi T = C\times(1-T_C/T)^{-d\nu} = C\times(1-T_C/T)^{-\gamma} \quad \text{for } T_C>0, \tag{4a}$$

$$\chi T = C\times\exp(d\Theta/T) = C\times\exp(W/T) \quad \text{for } T_C=0, \tag{4b}$$

$$\begin{aligned}\chi T &= C\times(1+T_K/T)^{d\Theta/T_K}\\ &= C\times(1+T_K/T)^{-\gamma} \quad \text{for } T_C=-T_K<0.\end{aligned} \tag{4c}$$

Solution (4a) looks like the familiar power law generally

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TABLE I. Critical exponent of the $\chi T = f(T)$ dependence, according to the spin space ($n$) and lattice ($d$) dimensionalities. The mean field value $\gamma=1$ is observed for all $n$ at $d \geqslant 4$. $\gamma$ diverges for each $n$, at a lower critical dimensionality, $d_c(n)$, which is a frontier between the solutions of Eqs. (4a) and (4c).

| | $n=1$ (Ising) | $n=2$ ($XY$) | $n=3$ (Hbg) | $n=\infty$ (sph.) |
|---|---|---|---|---|
| $d=1$ | $\infty$ | | $-1.25S$ | |
| $d=2$ | 1.75 | KT | $\infty$ | |
| $d=3$ | 1.25 | 1.32 | 1.387 | 2 |
| $d=4$ | 1 | 1 | 1 | 1 |
| Mean field | 1 | 1 | 1 | 1 |

used to describe the phase transitions.[7] Unlike the classical expression, it is appropriate to depict the magnetic behavior over the whole temperature range. The dependence of $\gamma$ on the spin and lattice dimensionalities is given in Table I, which describes a finite number of interesting cases, including two exact solutions, the Ising chain model ($n=1, d=1$) and the Onsager's solution for the 2D Ising model ($n=1, d=2$). It can be pointed out that the long-range order is destroyed if the space dimension is decreased below a lower critical dimensionality, $d_c(n)$, characterized by a divergence of $\gamma$.

Let us now focus upon the solutions of (4b) or (4c) that have, in general, been left aside. They have thermodynamically the same legitimacy as solution (4a), and therefore are candidates to describe systems where spin correlations exist, but no long-range order takes place at any finite temperature. For $T_C=0$, the finding coincides with the exact solution for the ferromagnetic Ising chain with nearest neighbor interactions.[16] We assume, more generally, that an exponential solution fits all situations, like the Ising chain, which corresponds to a lower critical dimensionality $d_c$. Conversely, we will show hereafter that the $T_C<0$ solution describes systems setting below a lower critical dimensionality, like the one-dimensional (1D) Heisenberg ferromagnets, and we suspect that it could be appropriate in all cases where $d \geqslant 1$.

Note that because we are using $J/T$, which cancels when $T$ diverges, rather than $T/J$ to construct the scaling variable $(J/T_C - J/T)$ all Eqs. (4) have a sensible high temperature expansion. Thus, the Curie-Weiss law $\chi = C/(T-W)$ is recovered, with $C=S(S+1)$ being the Curie constant (in $Ng^2\mu_B^2/3k$ unit), and $W=d\times\Theta$ the Weiss temperature.

In order to decide which expression is more appropriate to describe experiments, we have proposed to differentiate Eqs. (4) to deduce the equivalent expression

$$\partial \log(T)/\partial \log(\chi T) = (T - T_C)/\gamma T_C. \quad (5)$$

The plot of $\partial \log_{10}(T)/\partial \log_{10}(\chi T)$ vs $T$ gives a straight line, in the range where the model is valid, which intersects the temperature axis at a positive, null, or negative $T_C$ value, and the $T=0$ axis at $\gamma^{-1}$. In the $T_C=0$ limit, where $\chi T$ is described by Eq. (4b), the straight line intersects the axes at their origin. We show hereafter that such an approach furnishes the right framework to describe the ferromagnetic Heisenberg chains of quantum spins $S$, and to deduce an analytical expression of the magnetic susceptibility.

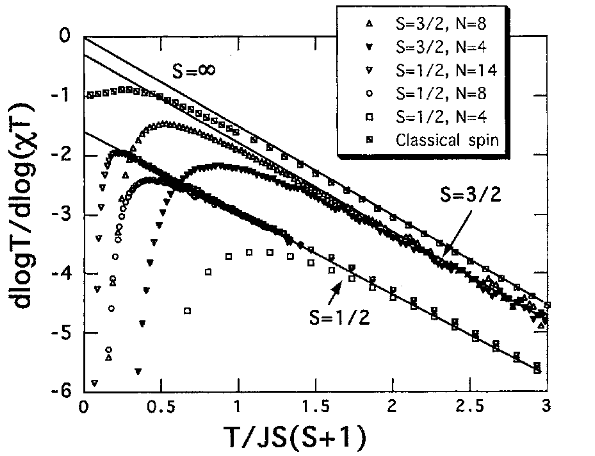


FIG. 1. Plot of the $\partial \log_{10}(T)/\partial \log_{10}(\chi T)$ function as determined by differentiating, numerically, the results of exact calculations of the susceptibility for ferromagnetic rings of $N$ Heisenberg spins $S=\frac{1}{2}$ and $\frac{3}{2}$ and for the classical limit $S=\infty$.

## III. DESCRIPTION OF FERROMAGNETIC HEISENBERG CHAINS

In what follows, we describe the magnetic behavior of finite ferromagnetic rings of $N$ Heisenberg spins ($S=\frac{1}{2}$, 1, $\frac{3}{2}$, etc.) by using scaling arguments developed in Sect. I. We have been guided by our confidence that: (i) superparamagnetic scaling should describe the infinite 1D-Heisenberg ferromagnetic chain, and (ii) the use of finite rings is allowed if the correlation length is much smaller than the size of the rings. It follows that superparamagnetic hierarchical scaling should be obeyed down to the lowest temperatures for very large rings. Thermodynamical data of the ferromagnetic rings, made of $N$ quantum spins $S$, were calculated by exact diagonalization of the spin Hamiltonian $H=-J\Sigma S_i S_{i+1}$, according to the procedure initiated by Bonner and Fisher for the $S=\frac{1}{2}$ AF chain,[17] and subsequently extended to arbitrary spin quantum numbers.[18–20] This well-documented numerical approach was used to get data with a good accuracy.

Figure 1 shows the temperature dependence of $\chi T$ as $\partial \log_{10}(T)/\partial \log_{10}(\chi T)$ plots for finite ferromagnetic rings of $N$ isotropic spins $S$. For a given $S$, $\partial \log_{10}(T)/\partial \log_{10}(\chi T)$ is a linear function of $T$, which is aiming a negative $T_C=-T_K$, down to a threshold value $T_S(N)$, the value of which decreases as $N$ increases. $T_S(N)$ corresponds to the maximum of $\partial \log_{10}(T)/\partial \log_{10}(\chi T)$. We observe that this linear part intercepts the axes at $T_C$ and $\gamma^{-1}$ (both negative) that stay much the same for different $N$ values. Only the linearity is better for larger $N$, so that Eq. (4c) better fits the data for larger $N$, suggesting the following expression of the susceptibility for the infinite chain:

$$\chi T = C \times (1 + \Theta JS(S+1)/T)^{-\gamma} \quad \text{with } \Theta > 0 \quad (6)$$

which holds for $T > T_S(\infty)$.

TABLE II. Best values of $C/[S(S+1)]$, $\gamma\Theta$, and $\gamma S$ for the 1D-Heisenberg ferromagnetic chains of spins $S=\frac{1}{2}$, 1, $\frac{3}{2}$. $C$, $\Theta$, and $\gamma$ (negative) are the parameters characterizing the scaling equation adapted to systems in a space dimension below a lower critical dimensionality, $\chi T=C[1+\Theta JS(S+1)/T]^{-\gamma}$.

| $S$ | $\frac{1}{2}$ | 1 | $\frac{3}{2}$ |
|---|---|---|---|
| $C/S(S+1)$ | 0.991 | 0.989 | 0.989 |
| $-\gamma\Theta$ | 0.742 | 0.735 | 0.731 |
| $-\gamma S$ | 1.222 | 1.232 | 1.164 |

Table II gives the best values of $C$, $\Theta$, and $\gamma$ for $S=\frac{1}{2},1,\frac{3}{2}$, obtained by fitting the susceptibility data for the largest rings, i.e., for $N=14$ $(S=\frac{1}{2})$, $N=10$ $(S=1)$ and $N=8$ $(S=\frac{3}{2})$, in the temperature range $T>T_S(N)$. In all cases, we observe that the Curie constant well agrees with the theoretical value $S(S+1)$, while $-\gamma\Theta$ is constant ($\sim$0.75) within 1%. The data display a negative curvature which signals a hierarchical scaling of the type of Eq. (4c), and that our model associates with systems below a lower critical dimensionality.

Finally we find, although with less accuracy (within 5%), that $\gamma=-1.25S$. These results enable us to propose the following general expression of $\chi T$ by using reduced temperature $T_R=T/S(S+1)$

$$\chi T_R=(1+0.6J/ST_R)^{1.25S} \quad \text{for } T_R>T_{R,S} \tag{7}$$

which is clearly more tractable than the polynomial expression reported for a given $S$ in the literature.[21,22] From this approach, we can deduce the very low temperature behavior of ferromagnetic chains, which is illustrated in Fig. 2 for $S=\frac{1}{2}$. Note that this result is directly comparable with the result of Baker *et al.* for $S=\frac{1}{2}$, obtained from Padé approximant technique.[22] If the series expansion is restricted to the first order in $1/T$, they obtain $\chi T\approx(1+0.75J/T)^{2/3}$, which is very similar to Eq. (7).

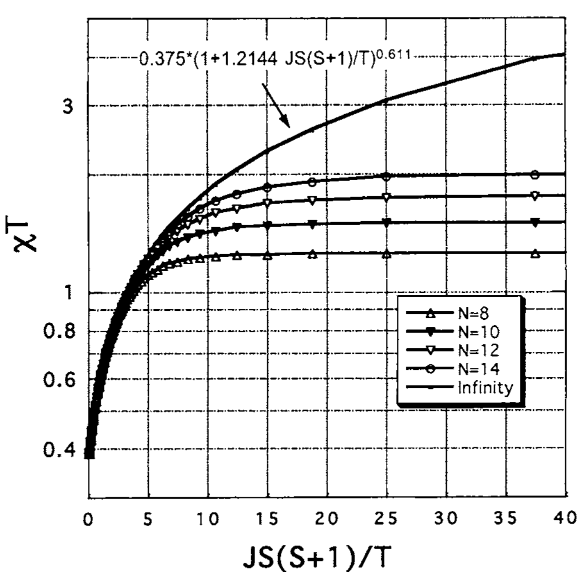


FIG. 2. Plot in Arrhenius coordinates of $\chi T(T)$, for finite ferromagnetic rings of $N$ Heisenberg spins $S=\frac{1}{2}$, and a prediction for the infinite Heisenberg chain.

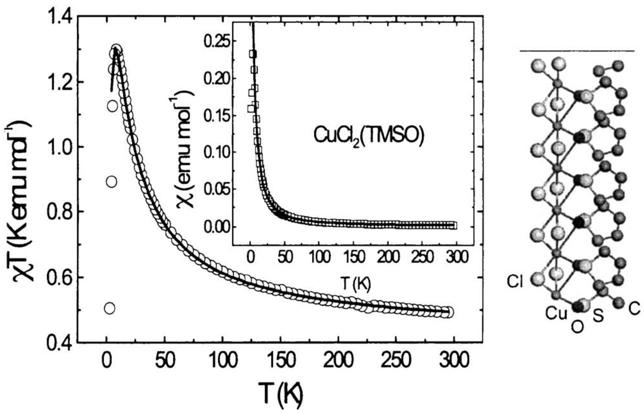


FIG. 3. View of the chain structure of $CuCl_2$(TMSO) and $\chi T=f(T)$ variation. The full line represents the best fit of the experimental data by using the power law expression (7) for $S=\frac{1}{2}$, corrected from small interchain interactions (see Ref. 27).

In the limit of classical spins ($S\rightarrow\infty$), expression (7) may be written

$$\chi T_R=\exp(0.75J/T_R) \quad \text{for } T_R>T_{R,S=\infty} \tag{8}$$

which indeed agrees with the theoretical expression of the susceptibility for $T$ larger than $JS(S+1)$.[23] According to our definition, therefore, $d=1$ may be viewed as a lower critical dimensionality for classical Heisenberg spins but not for quantum Heisenberg ones. The latter would belong to the space dimension below $d_C$ (Table I), since their magnetic susceptibility exhibits at low temperature a finite power law divergence, $\chi T\propto T^{-1.25S}$.

### A. Application to the ferromagnetic $S=\frac{1}{2}$ chain

The above model has been used for describing the behavior of the chain compound $CuCl_2$(TMSO), (TMSO is tetramethylsulfoxide) which is the archetype of the $S=\frac{1}{2}$. Heisenberg ferromagnetic chain.[24] The structure (Fig. 3) consists of the stacking of Cu(II) chains running along the b axis, in which copper(II) spin carriers interact through two chloro pathways and one oxo bridge. The chains are well separated in the space, the shortest Cu-Cu distance between neighboring chains being 7.2 Å.

The magnetic behavior of $CuCl_2$(TMSO),[25] shown in Fig. 3, exhibits a striking divergence of $\chi T(T)$ upon cooling down to 8 K, which is characteristic of a ferromagnetic chain behavior. The drop of $\chi T$ at lower temperature is ascribed to small AF interchain interactions, which may be of dipolar origin.[26] In order to describe the whole behavior, we used the expression of the susceptibility deduced from the scaling approach, namely, $\chi T=(g^2/8)S(S+1)[1+0.6J(S+1)/T]^{1.25S}$ with $S=\frac{1}{2}$. The effect of small interchain interactions ($zj$) has been introduced as a second order perturbation.[27]

Figure 3 shows that the magnetic susceptibility is perfectly fitted over the whole temperature range for $J=+93.55$ K, $zj=1.737$ K, and $g=2.12$, which agree with the values reported by Swank *et al.*[24] ($J=+74$ K, $zj=1.7$ K, and $g=2.09$) from the high temperature series expansion model. Notice that the latter is only valid for $T>J$, unlike the proposed expression which enables an accurate description of the properties at least down to $T/J=0.10$.

## IV. CONCLUSION

We have illustrated in this paper the pertinence of a strategy that extends to the description of correlated systems, the powerful ideas of scaling previously reserved to the sole phase transitions. On the basis of the sign and magnitude of the parameters characterizing the scaling, it is possible to extend the notion of universality class to these systems that stand at or below a lower critical dimensionality. This has been illustrated for the Heisenberg ferromagnetic chain, the $\chi T$ product of which is well described by a power law with a negative critical exponent, $-1.25S$, and a negative critical temperature.

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