Low-Temperature Study of Magnetic Properties of Two-Dimensional (2*d*) Classical Square Heisenberg Lattices

Jacques Curély
Laboratoire Ondes et Matière d'Aquitaine (LOMA)
Université Bordeaux 1 351 Cours de la Libération, 33405 Talence Cedex, France
E-mail: j.curely@loma.u-bordeaux1.fr

***Abstract*- We start this article by recalling rigorous results previously obtained for 2*d* square lattices composed of $(2N+1)^2$ classical spins isotropically coupled between first-nearest neighbors (i.e., showing Heisenberg couplings), in the thermodynamic limit ($N\to+\infty$) [4]: (i) the zero-field partition function $Z_N(0)$, (ii) the spin correlation (which vanishes in the zero-field limit, except at T=0 K so that the critical temperature is T_C=0 K, in agreement with Mermin-Wagner's theorem, (iii) the spin-spin correlation between any two lattice sites, (iv) the correlation length and (v) the static susceptibility. We exclusively focus on the low-temperature behaviors of the correlation length and the static susceptibility. This leads to the determination of a diagram characterized by three magnetic phases. Moreover we show that all the behaviors are in perfect agreement with the corresponding ones derived by using a renormalization method. Finally we give criterions allowing to directly determine the magnetic phases characterizing 2*d* magnetic compounds described by our microscopic model. An experimental test is given for illustrating this theoretical study.**

I. Introduction

Since the discovery of high-temperature superconductors, the nonlinear σ model in 2+1 dimensions has known a new interest because it allows the description of the properties of two-dimensional (2*d*) quantum antiferromagnets such as La_2CuO_4 [1]. Indeed these antiferromagnets, when properly doped, become superconductors up to a critical temperature T_C notably high compared to other types of superconducting materials. Under these conditions the nonlinear σ model has been conjectured to be equivalent at low temperature to the 2*d* Heisenberg model [2]. In addition, while considering the connection between the σ model and the spin ½ Heisenberg model on a square lattice, Chakravarty *et al.* [2a] have shown that the long-wave length, low-energy properties are well-described by a mapping to a 2*d*-classical Heisenberg magnet because all the effects of quantum fluctuations can be resorbed by means of adapted renormalizations of the coupling constants. This is the reason for which the study of magnetic properties in two dimensions is so important because these properties are always present: the static properties give a good image of the spin arrangement and thus remain a good starting point for the study of dynamic properties.

From a practical point of view, thin magnetic layers play an important role in many areas of technology. Namely, in the field of spintronics, these layers may appear at the interface between different semiconductors showing or not magnetic properties. In addition they constitute an intermediate step for building up 3*d* artificial magnets whose structure may be imposed (like for magnetic grains used in nanotechnologies) and are characterized by local spins of high quantum number.

In previous papers [3] we gave a treatment for the 2*d* square lattice composed of classical spins (for instance, ions Mn^{2+} and Fe^{3+} characterized by a spin quantum number equal to 5/2) and isotropically coupled between nearest neighbors (i.e., showing Heisenberg couplings). They were based upon an approximation that we shall recall. More recently, we published a couple of papers [4] in which we rigorously established, for the first time, the closed-form expressions of the zero-field partition function and the spin-spin correlation, respectively. In this article, after recalling these important results we study the low-temperature behaviors of the correlation length and the static susceptibility derived from the spin-spin correlation in the physical case (i.e., for infinite lattices). This leads to the determination of a diagram characterized by three magnetic phases. Moreover we show that all the behaviors are in perfect agreement with the corresponding ones derived by using a renormalization method. Finally we give criterions allowing to directly determine the magnetic phases characterizing 2*d* magnetic compounds described by isotropic (Heisenberg) interactions between nearest-spin neighbors. An experimental test is given for illustrating this study.

II. Theoretical Background

1. Generalities

We start from the general Hamiltonian describing a lattice characterized by a square unit cell and wrapped on a torus so that it contains $(2N+1)^2$ sites, each one being the carrier of a classical spin $\boldsymbol{S}_{i,j}$:

$$H=\sum_{i=-N}^{N}\sum_{j=-N}^{N}\left(H_{i,j}^{ex}+H_{i,j}^{mag}\right), \tag{1}$$

with:

$$H_{i,j}^{ex}=(J_1\boldsymbol{S}_{i,j+1}+J_2\boldsymbol{S}_{i+1,j})\boldsymbol{S}_{i,j}\ , \tag{2}$$

$$H_{i,j}^{mag}=-G_{i,j}S_{i,j}^{z}B\ , \tag{3}$$

where:

$$G_{i,j} = G \quad \text{if } i+j \text{ is even or zero,}$$

$$G_{i,j} = G' \quad \text{if } i+j \text{ is odd.} \tag{4}$$

In (2) we recall that J_1 and J_2 refer to the exchange interaction between nearest neighbors belonging to the horizontal lines and vertical rows of the lattice, respectively. In addition $J_i>0$ (respectively, $J_i<0$), with i=1,2, denotes an antiferromagnetic (respectively, ferromagnetic) coupling. $G_{i,j}$ is the Landé factor characterizing each spin $\boldsymbol{S}_{i,j}$ and expressed in $\mu_B/\hbar$ unit, where μ_B is Bohr's magneton and $\hbar=h/2\pi$, h being Planck's constant. *Now one may wonder why we consider a lattice wrapped on a torus.* At first sight, one can guess that it is due to the fact that a torus possesses more symmetry elements than a plane lattice. But, *in the infinite limit* ($N\rightarrow+\infty$), we previously showed [4a] that *these two types of lattices are characterized by the same partition function* Z_N. As a result, *all the thermodynamic functions derived from* Z_N *are similar*.

The field-dependent partition function $Z_N(B)$ is defined as:

$$Z_N(B) = \int d\boldsymbol{S}_{-N,-N} \cdots \int d\boldsymbol{S}_{i,j} \cdots \int d\boldsymbol{S}_{N,N} \exp\left(-\beta \sum_{i=-N}^{N} \sum_{j=-N}^{N} \left(H_{i,j}^{ex} + H_{i,j}^{mag}\right)\right), \tag{5}$$

where $\beta=1/k_BT$ is the Boltzmann factor (which must not be confused with the critical exponent β_C). At this step it must be noticed that the calculation of the field-dependent partition function $Z_N(B)$ is plainly more complicated because of the presence of the further term $H_{i,j}^{mag}$ in the exponential argument, for each site (i,j). This aspect will not be examined in the present article. Under these conditions, the zero-field partition $Z_N(0)$ is simply obtained by integrating the operator $\exp(-\beta H^{ex})$ over all the angular variables characterizing the states of all the classical spins belonging to the lattice.

Thus we may respectively define the susceptibility per lattice site and the correlation length as:

$$\chi_{i,j} = \beta \sum_k \sum_{k'} G_{i,j} G_{i+k,j+k'} \Gamma_{k,k'}, \tag{6}$$

$$\xi = \left(\frac{\sum_k \sum_{k'} (k^2 + k'^2) \left|\Gamma_{k,k'}\right|}{\sum_k \sum_{k'} \left|\Gamma_{k,k'}\right|} \right)^{1/2}. \tag{7}$$

where $\Gamma_{k,k'}$ is the *correlation function*:

$$\Gamma_{k,k'} = <\boldsymbol{S}_{i,j} . \boldsymbol{S}_{i+k,j+k'}> - <\boldsymbol{S}_{i,j}><\boldsymbol{S}_{i+k,j+k'}>. \tag{8}$$

In the previous equation, the bracket notation <…> means that we deal with a thermodynamic average. In other words, if we consider a lattice wrapped on a torus, characterized by a square unit cell and composed of $(2N+1)^2$ sites, each one being the carrier of a classical spin $\boldsymbol{S}_{i,j}$, we may define the spin-spin correlation between any two spins as:

$$<\boldsymbol{S}_{i,j} . \boldsymbol{S}_{i+k,j+k'}> = \frac{1}{Z_N(0)} \int d\boldsymbol{S}_{-N,-N} \cdots \int \boldsymbol{S}_{i,j} d\boldsymbol{S}_{i,j} \cdots \int \boldsymbol{S}_{i+k,j+k'} d\boldsymbol{S}_{i+k,j+k'} \cdots \int d\boldsymbol{S}_{N,N} \exp\left(-\beta \sum_{i=-N}^{N} \sum_{j=-N}^{N} H_{i,j}^{ex}\right) \tag{9}$$

where $Z_N(0)$ is the zero-field partition function given by (5) expressed in the zero-field limit.

2. *Recall of the calculation of $Z_N(0)$* [4a]

Because of the presence of classical spin moments, all the operators $H_{i,j}^{ex}$ commute and the exponential factor appearing in the integrand of (5) considered in the zero-field limit may be written:

$$\exp\left(-\beta \sum_{i=-N}^{N} \sum_{j=-N}^{N} H_{i,j}^{ex}\right) = \prod_{i=-N}^{N} \prod_{j=-N}^{N} \exp\left(-\beta H_{i,j}^{ex}\right). \tag{10}$$

As a result, the particular nature of $H_{i,j}^{ex}$ given by (2) allows one to separate the contributions corresponding to the exchange coupling involving classical spins belonging to the same horizontal line i of the layer (i.e., $\boldsymbol{S}_{i,j-1}$, $\boldsymbol{S}_{i,j+1}$ and $\boldsymbol{S}_{i,j}$) or to the same vertical row j (i.e., $\boldsymbol{S}_{i-1,j}$, $\boldsymbol{S}_{i+1,j}$ and $\boldsymbol{S}_{i,j}$). In fact, for each of the four contributions (one per bond connected to the site (i,j) carrying the spin $\boldsymbol{S}_{i,j}$), we have to expand a term such as $\exp(-A\boldsymbol{S}_1.\boldsymbol{S}_2)$ where A is βJ_1 or βJ_2 (the classical spins $\boldsymbol{S}_1$ and $\boldsymbol{S}_2$ being considered as unit vectors). If we call $\Theta_{1,2}$ the angle between vectors $\boldsymbol{S}_1$ and $\boldsymbol{S}_2$, respectively characterized by the couples of angular variables (θ_1,φ_1) and (θ_2,φ_2), it is possible to expand the operator $\exp(-A\cos\Theta_{1,2})$ on the infinite basis of spherical harmonics which are eigenfunctions of the angular part of the Laplacian operator on the sphere of unit radius S^2:

$$\exp\left(-A\cos\Theta_{1,2}\right) = 4\pi \sum_{\ell=0}^{+\infty} \left(\frac{\pi}{2A}\right)^{1/2} \mathrm{I}_{\ell+1/2}(-A) \sum_{m=-\ell}^{+\ell} Y_{\ell,m}^{*}(\boldsymbol{S}_1) Y_{\ell,m}(\boldsymbol{S}_2). \tag{11}$$

In the previous equation the $\mathrm{I}_{\ell+1/2}(-A)$'s are modified Bessel functions of the first kind and $\boldsymbol{S}_1$ and $\boldsymbol{S}_2$ symbolically represent the couples (θ_1,φ_1) and (θ_2,φ_2). If we set:

$$\lambda_\ell(-\beta j) = \left(\frac{\pi}{2\beta j}\right)^{1/2} \mathrm{I}_{\ell+1/2}(-\beta j), \; j = J_1 \text{ or } J_2, \tag{12}$$

each operator $\exp\left(-\beta H_{i,j}^{ex}\right)$ is finally expanded on the basis of eigenfunctions (the spherical harmonics), whereas the λ_ℓ's are nothing but the associated eigenvalues. Under these conditions, the zero-field partition function $Z_N(0)$ directly appears as a characteristic polynomial and may be written as:

$$Z_N(0)=(4\pi)^{2(2N+1)^2}\sum_{\ell_{N,-N}=0}^{+\infty}\lambda_{\ell_{N,-N}}(-\beta J_1)\sum_{\ell'_{N,-N}=0}^{+\infty}\lambda_{\ell'_{N,-N}}(-\beta J_2)\times\ldots$$

$$\times\sum_{\ell_{-N,N-1}=0}^{+\infty}\lambda_{\ell_{-N,N-1}}(-\beta J_1)\sum_{m_{N,-N}=-\ell_{N,-N}}^{+\ell_{N,-N}}\;\sum_{m'_{N,-N}=-\ell'_{N,-N}}^{+\ell'_{N,-N}}\ldots\sum_{m_{-N,N-1}=-\ell_{-N,N-1}}^{+\ell_{-N,N-1}}\;\prod_{i=-N}^{N}\prod_{j=-N}^{N}F_{i,j}\,, \tag{13}$$

$$F_{i,j}=\int d\boldsymbol{S}_{i,j}\,Y_{\ell'_{i+1,j},m'_{i+1,j}}(\boldsymbol{S}_{i,j})\,Y_{\ell_{i,j-1},m_{i,j-1}}(\boldsymbol{S}_{i,j})\,Y^{*}_{\ell_{i,j},m_{i,j}}(\boldsymbol{S}_{i,j})\,Y^{*}_{\ell'_{i,j},m'_{i,j}}(\boldsymbol{S}_{i,j}) \tag{14}$$

where $F_{i,j}$ is the current integral per site (with one spherical harmonic per bond). Using the decomposition of any pair of spherical harmonics [5], $F_{i,j}$ may be expressed as a Clebsch-Gordan (C.G.) series. Whatever the finite or infinite lattice size, the non-vanishing condition of each current integral $F_{i,j}$ is mainly due to that of C.G. coefficients. It allows one to derive two types of selection rules.

The *first selection rule* concerns the coefficients m and m' appearing in (14):

$$m_{i,j-1}+m'_{i+1,j}-m_{i,j}-m'_{i,j}=0. \tag{15}$$

The *second selection rule* is derived from the fact that the various coefficients ℓ and ℓ' appearing in (14) obey triangular inequalities [5]. It may be written under the two following relations:

$$\ell_{i,j-1}+\ell'_{i+1,j}+\ell_{i,j}+\ell'_{i,j}=2g_{i,j}, \tag{16a}$$

$$\ell_{i,j-1}+\ell'_{i+1,j}-\ell_{i,j}-\ell'_{i,j}=2g'_{i,j}, \tag{16b}$$

(or equivalently $\ell_{i,j}+\ell'_{i,j}-\ell_{i,j-1}-\ell'_{i+1,j}=2g''_{i,j}$, with $g''_{i,j}=-g'_{i,j}$) where $g_{i,j}$ or $g'_{i,j}$ is a relative integer.

Then, using (i) the invariance of $Z_N(0)$ under the permutation of indices 1 and 2, (ii) the symmetry elements of the lattice wrapped on a torus, it is easy to derive:

$$m_{i,j}=m'_{i,j}=m\;\;,\;\;\ell_{i,j}=\ell'_{i,j}=\ell\;\;\forall\,(i,j)\in\text{Torus}. \tag{17}$$

For a torus of infinite radius of curvature, we have shown that m=0. Under these conditions the zero-field partition function may be written in the *thermodynamic limit*:

$$Z_N(0)=(4\pi)^{8N^2}\sum_{\ell=0}^{+\infty}\left[F_{\ell,\ell}\lambda_\ell(-\beta J_1)\lambda_\ell(-\beta J_2)\right]^{4N^2},\quad \text{as } N\to+\infty \tag{18}$$

where the integral $F_{\ell,\ell}$ is the new simplified expression of integral $F_{i,j}$ (the factor $1/4\pi$ has been omitted):

$$F_{\ell,\ell}=(2\ell+1)^2\sum_{L=0}^{2\ell}\frac{1}{2L+1}\left[C_{\ell\;0\;\ell\;0}^{L\;0}\right]^4,\;F_{0,0}=1. \tag{19}$$

In addition we have shown that the series must be truncated for a given temperature. If we define the ratio r_ℓ such as:

$$r_\ell=\frac{\lambda_\ell(-\beta J_1)\lambda_\ell(-\beta J_2)}{\lambda_0(-\beta J_1)\lambda_0(-\beta J_2)}\frac{F_{\ell,\ell}}{F_{0,0}} \tag{20}$$

we may achieve a numerical study restricted to the case J=J_1=J_2 for sake of simplicity but the reasoning may be easily extended to the general case $J_1 \neq J_2$. *This is due to the fact that the expression of $Z_N(0)$ appears at the denominator of all the thermodynamic functions derived from $Z_N(0)$* such as the spin correlation, the correlation length and the static susceptibility.

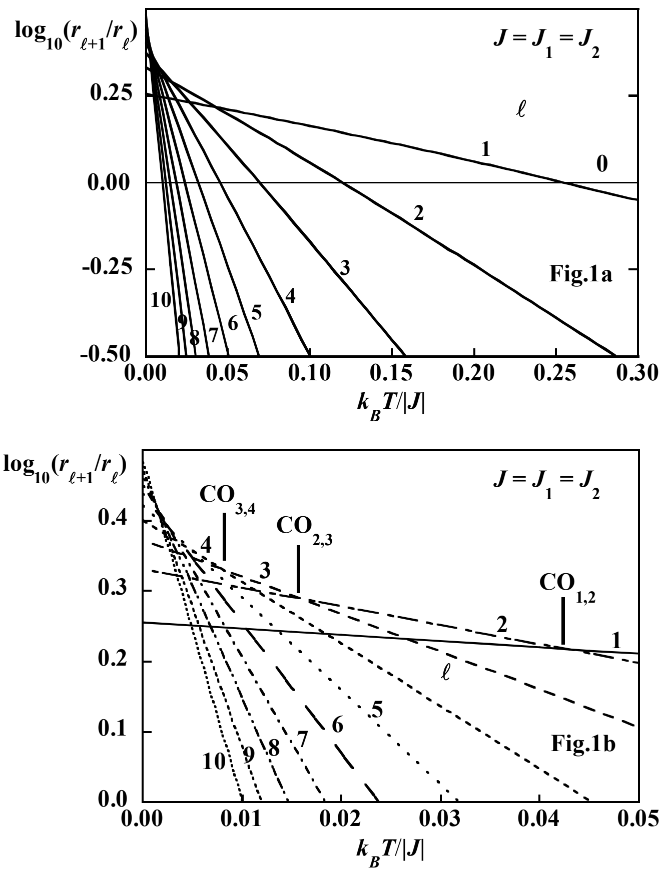

Fig. 1. a) Thermal variations of the ratio $\log_{10}(r_{\ell+1}/r_\ell)$ for various values of ℓ where r_ℓ is defined by (20); b) zoom of the plot allowing to have a better insight of the crossover phenomena between various ℓ-regimes.

If we zoom Fig.1a in the very low-temperature domain, we may then observe a succession of *crossovers* (see Fig.1b), each crossover being characterized by a specific temperature called *crossover temperature* T_{CO}. For instance, for the reduced temperature $k_BT/|J|\geq 0.255$, $\lambda_0(-\beta J)$ appears as the dominant term of the characteristic polynomial whereas, for $0.255\geq k_BT/|J|\geq 0.043$, $\lambda_1(-\beta J)$ becomes the dominant one etc... In that case the crossover temperature corresponding to the transition between the regimes respectively characterized by $\ell=0$ and $\ell=1$ is labelled $T_{CO_{0,1}}$. Note that, for $T=0$ K rigorously, all the eigenvalues intervene due to the fact that the crossover temperatures are closer and closer. The value $\ell\to+\infty$ becomes dominant. As a result we may say that $T=0$ K *plays the role of critical temperature*. We shall see that, in the low-temperature regime, this study will play an important role for determining a magnetic phase and the corresponding temperature law to which the spin-spin correlation obeys.

3. Spin correlations, correlation length and static susceptibility

$<\boldsymbol{S}_{i,j}.\boldsymbol{S}_{i+k,j+k'}>$ is called the *spin-spin correlation* whereas $<\boldsymbol{S}_u>$, with $u=(i,j)$ or $(i+k,j+k')$, is the *spin correlation.* In addition, as we deal with isotropic (Heisenberg) couplings, we have the following properties:

$$<S^{\nu}_{i,j}.S^{\nu}_{i+k,j+k'}> \; = \frac{1}{3}<\boldsymbol{S}_{i,j}.\boldsymbol{S}_{i+k,j+k'}>, \quad \nu = x, y \text{ or } z,$$

$$<S^{\nu}_u> = \frac{1}{3}<\boldsymbol{S}_u>,$$

$$\boldsymbol{S}_u = S^x_u\boldsymbol{e}_x + S^y_u\boldsymbol{e}_y + S^z_u\boldsymbol{e}_z, \; u = (i,j) \text{ or } (i+k,j+k'), \quad (21)$$

from which we immediately derive for the correlation function:

$$\Gamma^{\nu}_{k,k'} = \frac{1}{3}\Gamma_{k,k'}, \quad \nu = x, y \text{ or } z. \quad (22)$$

Finally we may define the *self spin-spin correlation* $<(S^{\nu}_u)^2>$, with $\nu=x$, y or z. We have $<\boldsymbol{S}^2>=1$ due to the fact that the classical spin is considered as a unit vector. Consequently, as we deal with isotropic spin-spin couplings, we may write:

$$<(S^x_u)^2> = <(S^y_u)^2> = <(S^z_u)^2> = \frac{1}{3},$$

$$\chi^x_{i,j} = \chi^y_{i,j} = \chi^z_{i,j} = \frac{\chi_{i,j}}{3}, \; \xi^x = \xi^y = \xi^z = \xi = \frac{\xi}{\sqrt{3}}. \quad (23)$$

The calculation of the spin correlation $<S^{\nu}_u>$ and the spin-spin correlation $<S^{\nu}_{i,j}.S^{\nu}_{i+k,j+k'}>$ is strictly similar to that of the zero-field partition function $Z_N(0)$. We have found:

$$<S^z_{i,j}> = 0, \; <S^z_{i+k,j+k'}> = 0,$$

$$\Gamma^z_{k,k'} = \; <S^z_{i,j}.S^z_{i+k,j+k'}> \text{ for } T > 0 \text{ K}. \quad (24)$$

Of course, when $T=0$ K exactly, we have: $|<S^z_{i,j}>|=1$. *This result rigorously proves that the critical temperature is absolute zero* i.e., $T_C=0$ K.

The spin-spin correlation in the thermodynamic limit ($N\to+\infty$) is given by [4b]:

$$<S^z_{0,0}.S^z_{k,k'}> = \frac{1}{Z_N(0)}\sum_{\ell=0}^{+\infty}\left[F_{\ell,\ell}\lambda_\ell(-\beta J_1)\lambda_\ell(-\beta J_2)\right]^{4N^2}\left[X_{\ell+1}+(1-\delta_{\ell,0})X_{\ell-1}\right], \; k>0, k'>0, \text{ as } N\to+\infty \quad (25)$$

where $Z_N(0)$ is the zero-field partition function given by (18), $\delta_{\ell,0}$ is the Dirac function and with:

$$X_{\ell+\varepsilon} = \left(C_{\ell+\varepsilon}\right)^2\frac{F_{\ell,\ell+\varepsilon}}{F_{\ell,\ell}}\left(u_{1,\ell+\varepsilon}\right)^{k'}\left(u_{2,\ell+\varepsilon}\right)^{k}, \; k\geq 0, k'\geq 0, \varepsilon = \pm 1, \; C_{\ell+1} = \frac{\ell+1}{\sqrt{(2\ell+1)(2\ell+3)}}, \; C_{\ell-1} = \frac{\ell}{\sqrt{(2\ell+1)(2\ell-1)}}, \quad (26a)$$

$$u_{i,\ell+\varepsilon} = \frac{F_{\ell,\ell+\varepsilon}}{F_{\ell,\ell}}\frac{\lambda_{\ell+\varepsilon}(-\beta J_i)}{\lambda_\ell(-\beta J_i)}, \; \varepsilon = \pm 1, \; i = 1, 2. \quad (26b)$$

Integral $F_{\ell,\ell}$ is given by (19) and integral $F_{\ell,\ell+\varepsilon}$ is:

$$F_{\ell,\ell+\varepsilon} = (2\ell+1)(2\ell+2\varepsilon+1)\sum_{L=0}^{\min(2\ell,2\ell+2\varepsilon)}\frac{1}{2L+1}\left[C^{L\;0}_{\ell\;0\;\ell\;0}C^{L\;0}_{\ell+\varepsilon\;0\;\ell+\varepsilon\;0}\right]^2, F_{0,\varepsilon} = 1, \; \varepsilon = \pm 1. \quad (27)$$

The correlation length may be derived from (7) and (25)-(27):

$$\xi = \left[\frac{\sum_{\ell=0}^{+\infty}\left[F_{\ell,\ell}\lambda_\ell(-\beta J_1)\lambda_\ell(-\beta J_2)\right]^{4N^2}\left[N_{\ell+1}+(1-\delta_{\ell,0})N_{\ell-1}\right]}{\sum_{\ell=0}^{+\infty}\left[F_{\ell,\ell}\lambda_\ell(-\beta J_1)\lambda_\ell(-\beta J_2)\right]^{4N^2}\left[D_{\ell+1}+(1-\delta_{\ell,0})D_{\ell-1}\right]}\right]^{1/2}, \text{ as } N\to+\infty, \quad (28)$$

with:

$$D_{\ell+\varepsilon} = (C_{\ell+\varepsilon})^2 \frac{F_{\ell,\ell+\varepsilon}}{F_{\ell,\ell}} \frac{1}{(1-|u_{1,\ell+\varepsilon}|)(1-|u_{2,\ell+\varepsilon}|)}, \; N_{\ell+\varepsilon} = D_{\ell+\varepsilon}\left[\frac{|u_{1,\ell+\varepsilon}|(1+|u_{1,\ell+\varepsilon}|)}{(1-|u_{1,\ell+\varepsilon}|)^2} + \frac{|u_{2,\ell+\varepsilon}|(1+|u_{2,\ell+\varepsilon}|)}{(1-|u_{2,\ell+\varepsilon}|)^2}\right]. \quad (29)$$

where $C_{\ell+\varepsilon}$ and $u_{i,\ell+\varepsilon}$ are given by (26a) and (26b), respectively as well integrals $F_{\ell,\ell+\varepsilon}$ and $F_{\ell,\ell}$ by (19) and (27).

The static susceptibility per square unit cell and averaged per site may be written as:

$$\chi = \frac{1}{4}\left(\chi^z_{0,0} + \chi^z_{0,1} + \chi^z_{1,0} + \chi^z_{1,1}\right) \quad (30)$$

where the susceptibility per site is given by:

$$\chi_{k,k'} = \frac{1}{Z_N(0)} \sum_{\ell=0}^{+\infty} \left[F_{\ell,\ell}\lambda_\ell(-\beta J_1)\lambda_\ell(-\beta J_2)\right]^{4N^2} \left[\chi_{\ell+1} + (1-\delta_{\ell,0})\chi_{\ell-1}\right], \text{ as } N\to+\infty \; (k = 0 \text{ or } 1,\, k' = 0 \text{ or } 1) \quad (31)$$

with:

$$\chi_{\ell+\varepsilon} = \frac{\beta}{6}(C_{\ell+\varepsilon})^2 \frac{F_{\ell,\ell+\varepsilon}}{F_{\ell,\ell}} \frac{(G^2 + G'^2)W_{1,\ell+\varepsilon} + 2GG'W_{2,\ell+\varepsilon}}{W_{3,\ell+\varepsilon}}, \; \varepsilon = \pm 1, \quad (32)$$

$$W_{1,\ell+\varepsilon} = [1+(u_{1,\ell+\varepsilon})^2][1+(u_{2,\ell+\varepsilon})^2] + 4u_{1,\ell+\varepsilon}u_{2,\ell+\varepsilon}, \; W_{2,\ell+\varepsilon} = u_{1,\ell+\varepsilon}(1+(u_{2,\ell+\varepsilon})^2) + u_{2,\ell+\varepsilon}(1+(u_{1,\ell+\varepsilon})^2), \quad (33)$$

$$W_{3,\ell+\varepsilon} = [1-(u_{1,\ell+\varepsilon})^2][1-(u_{2,\ell+\varepsilon})^2], \quad (34)$$

where $C_{\ell+\varepsilon}$ and $u_{i,\ell+\varepsilon}$ are given by (26a) and (26b), respectively as well integrals $F_{\ell,\ell}$ and $F_{\ell,\ell+\varepsilon}$ by (19) and (27).

III. LOW-TEMPERATURE STUDY

The low-temperature study of the correlation length and the static susceptibility may be achieved by remarking that these thermodynamic functions are respectively characterized by a denominator of the type $1-|u_{i,\ell+\varepsilon}|$ or $1-(u_{i,\ell+\varepsilon})^2$, with i=1, 2, where $u_{i,\ell+\varepsilon}$ is given by (26b). As a result the study of the ratio $u_{i,\ell+\varepsilon}$ (i.e., the study of ratios $\lambda_{\ell+\varepsilon}(-\beta|J_i|)/\lambda_\ell(-\beta|J_i|)$ and $F_{\ell,\ell+\varepsilon}/F_{\ell,\ell}$) is going to be crucial. In addition we have observed that, in the low-temperature regime, we must consider $\ell\to+\infty$ as well as the argument $\beta|J_i|$ (see Fig.1b).

1. Preliminaries

We first examine the ratio $F_{\ell,\ell+\varepsilon}/F_{\ell,\ell}$ ($\varepsilon=\pm1$) where integrals $F_{\ell,\ell}$ and $F_{\ell,\ell+\varepsilon}$ given by (19) and (27) have been evaluated as C.G. series. However, under this form, it is difficult to express their behaviour when $\ell\to+\infty$. For this reason, we prefer starting from their integral definition given by (14) and involving spherical harmonics. We have [5]:

$$Y_{\ell,0}(\theta,\varphi) \approx \frac{1}{\pi\sqrt{\sin\theta}}\left\{\left(1-\frac{3}{8\ell}\right)\cos\left((2\ell+1)\frac{\theta}{2}-\frac{\pi}{4}\right) - \frac{1}{8\ell\sin\theta}\cos\left((2\ell+3)\frac{\theta}{2}-\frac{3\pi}{4}\right)\right\} + o\left(\frac{1}{\ell^2}\right), \text{ as } \ell\to+\infty,$$

$$\varepsilon' \le \theta \le \pi-\varepsilon', \; 0<\varepsilon' \ll 1/\ell, \; 0 \le \varphi \le 2\pi. \quad (35)$$

Then, using this asymptotic behavior, a tedious but not complicated calculation allows one to show that:

$$F_{\ell,\ell} \approx \frac{1}{\pi^3}\ln(\ell), \text{ as } \ell \to +\infty, \quad (36)$$

so that:

$$\frac{F_{\ell,\ell+\varepsilon}}{F_{\ell,\ell}} \to 1 + \frac{\varepsilon}{\ell\ln(\ell)}, \; \varepsilon = \pm1, \text{ as } \ell \to +\infty. \quad (37)$$

This ratio is always slightly greater (ε=+1) or slightly lower (ε=−1) than unity and tends to unity by increasing ℓ-values.

Now we must examine the ratio $\lambda_{\ell+\varepsilon}(-\beta|J_i|)/\lambda_\ell(-\beta|J_i|)$ i.e., $I_{\ell+\varepsilon+1/2}(-\beta|J_i|)/I_{\ell+1/2}(-\beta|J_i|)$, with $\varepsilon=\pm1$, i=1,2, as $\ell\to+\infty$. Intuitively, in the low-temperature limit, we must consider the three cases $\beta|J_i|>>\ell$, $\beta|J_i|\sim\ell$ and $\beta|J_i|<<\ell$. The behavior of Bessel function $I_{\ell+1/2}(-\beta|J_i|) \approx I_\ell(-\beta|J_i|)$ as $\ell\to+\infty$ and $\beta|J_i|\to+\infty$ has been established by Olver [6a, 6b]. We have extended this work for a large order ℓ (but not necessarily infinite) and for any real argument z_i varying from a finite value to infinity. Thus, the study of the Bessel differential equation in the large ℓ-limit necessitates the introduction of the dimensionless auxiliary variables:

$$\zeta_i = -\frac{J_i}{|J_i|}\left[\sqrt{1+z_i^2} + \ln\left(\frac{|z_i|}{1+\sqrt{1+z_i^2}}\right)\right], \; |z_i| = \frac{\beta|J_i|}{\ell}. \quad (38)$$

The numerical study of $|\zeta|$ is reported in Fig.2. As expected we observe that there are two branches. $|\zeta|$ vanishes for a numerical value of $|z_0|^{-1}$ very closed to $\pi/2$ so that there are 3 domains which will be physically interpreted in next subsection. Let be T_0 the corresponding temperature; we set:

$$\ell\frac{k_B T_0}{|J_i|} = \frac{\pi}{2}, \; i = 1, 2. \quad (39)$$

In the formalism of renormalization group T_0 is called a *fixed point*. In the present $2d$ case we have $\ell\to+\infty$. As a result we

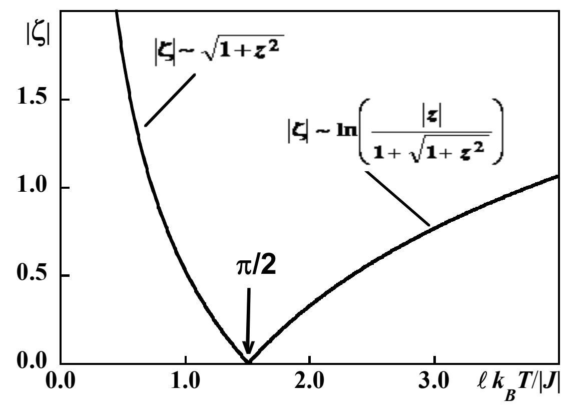

Fig. 2. Thermal variations of $|\zeta|$ for various values of $\ell k_B T/|J|$.

derive that $T_0 \to T_C=0$ K as $\ell \to +\infty$ so that the critical temperature may be seen as a fixed point. This will allow one to expand all the thermodynamic functions as series of current term $|T-T_0|$ near $T_0 \approx T_C=0$ K.

At this step we must recall that the spin modulus $S(S+1) \sim S^2$, as $S \to +\infty$, is absorbed in the exchange energy J. Chakravarty *et al.* [2a] as well as Chubukov *et al.* [2b] have written that the action $S/\hbar$ (which allows one to calculate the partition function) is proportional to $J/2$. In addition these authors have considered the spin density S/a where a is the lattice spacing. In our case the lattice spacing between two similar Landé factors G or G' is $2a$. As a result, the left member of (39) may be written as $\ell k_B T_0/(|J|/2)(S/2a)^2$ so that the right member $\pi/2$ becomes 4π. We must keep in mind this remark because it will be very useful later.

The solutions of the Bessel equation in the large ℓ-limit may then be written as:

$$w_{\substack{1\\2}}(\zeta_i) \approx \exp(\pm \ell \zeta_i) \sum_{s=0}^{+\infty} (\pm 1)^s \frac{U_s(\zeta_i)}{\ell^s}, \ i = 1, 2. \qquad (40)$$

with the sign + (respectively, −) for w_1 (respectively, w_2). The coefficients $U_s(\zeta)$ have been expressed by Olver owing to an integro-differential equation [6a]. We only give below the first coefficients (with i=1, 2) that we need:

$$U_0(\theta_i) = 1, \quad U_1(\theta_i) = \frac{\theta_i}{8} - \frac{5\theta_i^3}{24},$$

$$U_2(\theta_i) = \frac{9\theta_i^2}{128} - \frac{77\theta_i^4}{192} + \frac{385\theta_i^6}{1152}, \quad \theta_i = \frac{1}{\sqrt{1+z_i^2}}. \qquad (41)$$

As a result, when ℓ is large without being necessarily infinite [6a], the Bessel function $I_{\ell+1/2}(\ell|z_i|)$ appears as a linear combination of $w_1(\zeta_i)$ and $w_2(\zeta_i)$ so that we may write:

$$I_{\ell+1/2}\left(\ell|z_i|\right) \approx \frac{(1+z_i^2)^{-1/4}}{\sqrt{2\pi\ell}} \left\{ \exp\left(\ell|\zeta_i|\right) \sum_{s=0}^{+\infty} \frac{U_s(\theta_i)}{\ell^s} \right.$$

$$\left. + \exp\left(-\ell|\zeta_i|\right) \sum_{k=0}^{+\infty} (-1)^s \frac{U_s(\theta_i)}{\ell^s} \right\}, \ i = 1, 2 \qquad (42)$$

where ζ_i is given by (38). If we set

$$f_\ell(\ell z_i) = \left(\frac{\pi}{2\ell z_i}\right)^{1/2} I_{\ell+1/2}(\ell z_i), \ i = 1, 2, \qquad (43)$$

with the parity property:

$$f_\ell(\ell z_i) = \left(-\frac{J_i}{|J_i|}\right)^\ell f_\ell(\ell|z_i|), \quad |z_i| = \frac{\beta|J_i|}{\ell}, \ i = 1, 2, \qquad (44)$$

we must finally study the ratios $f_{\ell+1}(\ell z_i)/f_\ell(\ell z_i)$ and $f_{\ell-1}(\ell z_i)/f_\ell(\ell z_i)$. Using the well-known relations [6b]:

$$f_{\ell+1}(\ell|z_i|) = \frac{df_\ell(\ell|z_i|)}{d(\ell|z_i|)} - \frac{\ell}{\ell|z_i|} f_\ell(\ell|z_i|),$$

$$f_{\ell-1}(\ell|z_i|) - f_{\ell+1}(\ell|z_i|) = \frac{2\ell+1}{\ell|z_i|} f_\ell(\ell|z_i|), \ i = 1, 2, \qquad (45)$$

we finally derive:

$$\frac{f_{\ell\pm1}(\ell z_i)}{f_\ell(\ell z_i)} = -\frac{J_i}{|J_i|} \left\{ \mp \left(\frac{1}{|z_i|} + \frac{1}{2\ell|z_i|} \right) + \frac{I'_{\ell+1/2}(\ell|z_i|)}{I_{\ell+1/2}(\ell|z_i|)} \right\},$$

$$i = 1, 2, \qquad (46)$$

with $I'_{\ell+1/2}(\ell|z_i|)=dI_{\ell+1/2}(\ell|z_i|)/d(\ell|z_i|)$ and $I_{\ell+1/2}(\ell|z_i|) \approx I_\ell(\ell|z_i|)$ so that $I'_{\ell+1/2}(\ell|z_i|) \approx dI_\ell(\ell|z_i|)/d(\ell|z_i|)$. Note that, in (46), we take into account that ℓ may be large without being necessarily infinite. Derivating (42) with respect to $\ell|z_i|$ we obtain:

$$I'_{\ell+1/2}\left(\ell|z_i|\right) \approx \frac{(1+z_i^2)^{1/4}}{\sqrt{2\pi\ell}\,|z_i|} \left\{ \exp\left(\ell|\zeta_i|\right) \sum_{s=0}^{+\infty} \frac{V_s(\theta_i)}{\ell^s} \right.$$

$$\left. - \exp\left(-\ell|\zeta_i|\right) \sum_{k=0}^{+\infty} (-1)^s \frac{V_s(\theta_i)}{\ell^s} \right\}, \ i = 1, 2. \qquad (47)$$

The coefficients $V_s(\theta_i)$ are polynomials in θ_i given by:

$$V_s = U_s - \theta_i(1-\theta_i^2)\left(\frac{1}{2}U_{s-1} + \theta_i \frac{dU_{s-1}}{d\theta_i}\right), \ s \geq 1. \qquad (48)$$

The first three (with i=1, 2) are [6a,6b]:

$$V_0(\theta_i) = 1, \quad V_1(\theta_i) = -\frac{3\theta_i}{8} + \frac{7\theta_i^3}{24},$$

$$V_2(\theta_i) = -\frac{15\theta_i^2}{128} + \frac{99\theta_i^4}{192} - \frac{455\theta_i^6}{1152}. \qquad (49)$$

Under these conditions it now becomes possible to derive the ratios $f_{\ell+1}(\ell z_i)/f_\ell(\ell z_i)$ and $f_{\ell-1}(\ell z_i)/f_\ell(\ell z_i)$, when ℓ becoming large but not necessarily infinite, for a z-argument varying from a finite value to infinity. Reporting the corresponding result as well as the behavior of $F_{\ell,\ell+\varepsilon}/F_{\ell,\ell}$ (with $\varepsilon=\pm1$) in the large ℓ-limit (cf (37)), it is then possible to express the ratio $u_{i,\ell+\varepsilon}$ defined by (26b) and appearing in the denominator of the correlation length and the static susceptibility in the low-temperature range. As previously remarked few lines before (38), in the low-temperature limit, we must consider the three cases $\beta|J_i|>>\ell>>1$ ($|z_i|>>1$), $\beta|J_i|\sim\ell>>1$ ($|z_i|\sim1$) and $\ell>>\beta|J_i|>>1$ ($|z_i|<<1$). The variable $\theta_i=1/\sqrt{1+z_i^2}$ is always lower than unity so that it remains possible to achieve a polynomial division in (46) if expressing (42) and (47) in the zero-temperature limit. We then find:

$$\frac{f_{\ell\pm1}(\ell z_i)}{f_\ell(\ell z_i)}=-\frac{J_i}{|J_i|}\left\{\mp\left(\frac{1}{|z_i|}+\frac{1}{2\ell|z_i|}\right)+\frac{1}{\theta_i|z_i|}\left[1-\frac{\theta_i}{2\ell}\right.\right.$$
$$\left.\left.-2\left(1-\frac{\theta_i}{4\ell}\right)\exp\left(-2\ell|\zeta_i|\right)+...\right\},\ i=1,2. \quad (50)$$

Introducing (37) and (50) in (26b) allows one to express the key ratio $u_{i,\ell\pm1}=f_{\ell\pm1}(\ell z_i)/f_\ell(\ell z_i)$ in the low-temperature limit.

2. *Low-temperature behaviors*

We begin this study by noting that it becomes necessary to expand the dimensionless variable $|\zeta_i|$ as a Taylor series of current term $|z_i|-z_0$, i=1,2, in order to derive $\exp(-2\ell|\zeta_i|)$ near z_0. In this region we have $|z_i|$ close to $2/\pi<1$ and we must evaluate the various derivatives $d^n|\zeta|/d|z|^n$ when $|z_i|=z_0$. We start from (38) so that we may write

$$\frac{d|\zeta_i|}{d|z_i|}=\frac{\sqrt{1+z_i^2}}{|z_i|}\approx\frac{1}{|z_i|},\ \text{as } |z_i|<<1,\ \left.\frac{d|\zeta_i|}{d|z_i|}\right|_{|z_i|=|z_0|}\approx\frac{\pi}{2} \quad (51)$$

and:

$$\frac{1}{n!}\frac{d^n|\zeta_i|}{d|z_i|^n}\approx\frac{(-1)^{n-1}}{n}\frac{1}{|z_i|^n},\ \frac{1}{n!}\left.\frac{d^n|\zeta_i|}{d|z_i|^n}\right|_{|z_i|=|z_0|}\approx\frac{(-1)^{n-1}}{n}\left(\frac{\pi}{2}\right)^n. \quad (52)$$

Noting that

$$\left||z_i|-z_0\right|=\frac{1}{\ell}\frac{X_i}{T},\ X_i=\frac{|J_i|}{k_B}\frac{|T-T_0|}{T_0},\ X_i=\begin{cases}\rho_i, & T<T_0\\ \Delta_i, & T>T_0\end{cases}, \quad (53)$$

the Taylor series giving $|\zeta_i|$ becomes:

$$|\zeta_i|=\ln\left(1+\frac{\pi}{2\ell}\frac{X_i}{T}\right),\ i=1,2, \quad (54)$$

and we may write:

$$\exp\left(-2\ell|\zeta_i|\right)=\left(1+\frac{\pi}{2\ell}\frac{X_i}{T}\right)^{-2\ell},\ i=1,2. \quad (55)$$

Noting that $(1\pm u/\ell)^\ell=\exp(\pm u)$, as ℓ large (or infinite), we finally derive:

$$\exp\left(-2\ell|\zeta_i|\right)=\exp\left(-\pi\frac{X_i}{T}\right),\ i=1,2,\ \text{as } \ell\to+\infty. \quad (56)$$

The physical meaning of ρ_i and Δ_i will be given below.

At this step we define the coupling constants at temperature T and at the fixed point T_0:

$$g=\frac{k_BT}{|J|},\ g_0=\frac{k_BT_0}{|J|},\ \bar{g}=\frac{T}{T_0}. \quad (57)$$

These ratios measure the strength of the quantum fluctuations because, if we recall that $|J|$ must be read as $|J|S^2$, fluctuations are inversely proportional to S^2. If expanding $|\zeta|$ near T_0 we have the following equations:

$$|\zeta_F|_<=\frac{\pi}{2}(1-\bar{g}),\ (T<T_0),\ |\zeta_F|_>=\frac{\pi}{2}(\bar{g}-1),\ (T>T_0), \quad (58)$$

so that the thermal study of $|\zeta|$ is reduced to two domains $\bar{g}<1$ i.e., $g<g_0$ ($T<T_0$) and $\bar{g}>1$ i.e., $g>g_0$ ($T>T_0$). Each of these domains may be itself divided in two subdomains according to as $|\zeta|>|\zeta_F|$ or $|\zeta|<|\zeta_F|$. This is the reason for which we are led to introduce the following variables:

$$x_1^i=\frac{k_BT}{|J_i||\zeta_F|_<},\ x_2^i=\frac{k_BT}{|J_i||\zeta_F|_>} \quad (59)$$

i.e.,

$$x_1^i=\frac{2T}{\pi\rho_i},\ x_2^i=\frac{2T}{\pi\Delta_i}. \quad (60)$$

Thus, as noted by Chubukov *et al.* [2b], the parameters x_1^i and x_2^i control the scaling properties of the magnetic system. ρ_i represents the *spin stiffness* of the ordered ground state (Néel state for an antiferromagnet) and Δ_i is a *spin gap between the ground state and the first excited one*. ρ_i and Δ_i (*cf* (53)) vanish at the fixed point T_0. If we report to the definition of $|\zeta|$ (*cf* (38)) $|\zeta|$ is homogeneous to T_0/T i.e., $1/\bar{g}$. If $\bar{g}<1$ ($T<T_0$) we have two possibilities $x_1^i>1$ i.e., $T>|\zeta_F|_<$ and $x_1^i<1$ i.e., $T<|\zeta_F|_<$. When $T=T_0$, x_1^i and x_2^i become infinite due to the fact that ρ_i and Δ_i vanish. Finally, if $\bar{g}>1$ ($T>T_0$) we have two possibilities $x_2^i>1$ i.e., $T>|\zeta_F|_>$ and $x_2^i<1$ i.e., $T<|\zeta_F|_>$. As there is an analytical continuity between x_1^i and x_2^i while passing through T_0, we derive that there are 3 domains: i) if $T<|\zeta_F|_<$ ($x_1^i<1$) we deal with the *Renormalized Classical Regime* (RCR), ii) if $T<|\zeta_F|_>$ ($x_2^i<1$) we deal with the *Quantum Disordered Regime* (QDR) and iii) if $T>|\zeta_F|_<$ ($x_1^i>1$) or $T>|\zeta_F|_>$ ($x_2^i>1$) we have the *Quantum Critical Regime* (QCR). Along the line $T=T_0$ in this domain we direc-

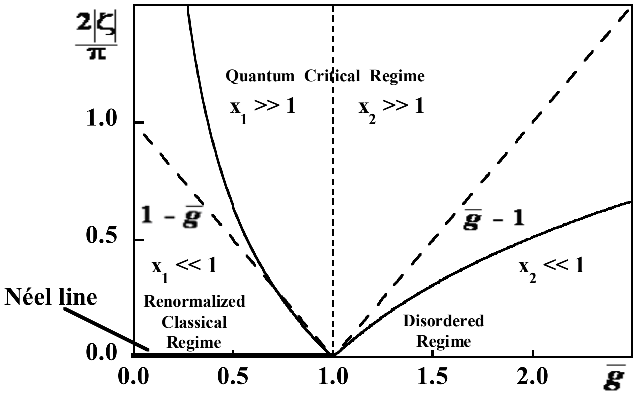

Fig. 3. Diagram of magnetic phases near $T_0 = T_C = 0$ K.

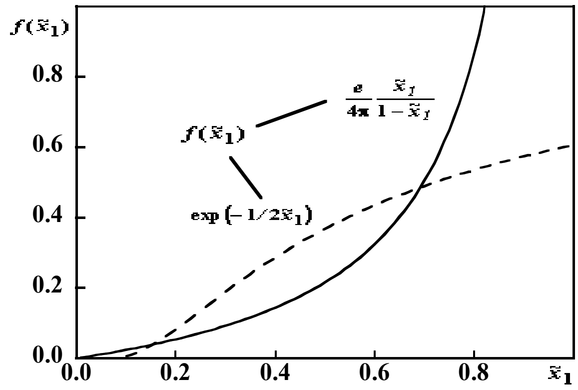

Fig. 4. Predominance of terms in the low-temperature expansion of the ratio $u_{i,\ell\pm1}=f_{\ell\pm1}(\ell z_i)/f_\ell(\ell z_i)$ given by (62) and (63).

tly reach the fixed point without a phase change. The magnetic diagram has been reported in Fig.3. It is absolutely similar to the one obtained by Chakravarty *et al.* [2a] and Chubukov *et al.* [2b], separately. Finally, if T tends towards absolute zero when coming from the *Renormalized Classical Regime* (RCR) or from the *Quantum Disordered Regime* (QDR), or directly tends to the fixed point T_0 when coming from the *Quantum Critical Regime* (QCR), we directly reach the *Néel line*. From now, due to the lack of place, we are going to restrict the low-temperature study to the case $T<T_0$.

We directly give below the key ratio $u_{i,\ell\pm1}=f_{\ell\pm1}(\ell z_i)/f_\ell(\ell z_i)$ in the low-temperature limit, for $T<T_0$, without detailing the calculations because they are long and hard (more than 12 pages of tight lines). As noted before (40) we use the conventional writing of Chakravarty *et al.* [2a] and Chubukov *et al.* [2b] for it allows one to make a reasoning with the simplest lattice unit cell of spacing a (instead of $2a$ in the most general case). As a result we shall reduce the physical discussion to the particular case $G=G'$. The ratio x_1^i now becomes $\widetilde{x}_1^i$ with the relationship $\widetilde{x}_1^i = x_1^i/4$. We then may write:

$$u_{i,\ell\pm1} = -\frac{J_i}{|J_i|}\left\{\mp\left(\frac{1}{|z_i|}+2\widetilde{x}_1^i\right)+1-2\widetilde{x}_1^i - \frac{8\pi}{e}\left(1-\widetilde{x}_1^i\right)\exp\left(-1/2\widetilde{x}_1^i\right)+\ldots\right\},\ \widetilde{x}_1^i<1,\ i=1,2. \quad (61a)$$

At this step we can note that the term $(1/|z_i|+2\,\widetilde{x}_1^i)$ may be momentarily omitted because the sum $u_{i,\ell+1}+u_{i,\ell-1}$ appears in the low-temperature expression of the spin-spin correlation and the correlation length. However this term must be refreshed for studying the susceptibility near 0 K. Using (53) and (60) allows one to write $\ell|z_i|=\ell|z_0|+1/x_1^i$ i.e., $\ell|z_i|=\ell|z_0|+4/\widetilde{x}_1^i$. Thus, when $\widetilde{x}_1^i>>1$, we have $\ell|z_i|\approx\ell|z_0|$ so that $1/\ell|z_i|\approx 1/\ell|z_0|$ vanishes as $\ell\to+\infty$ in (50). As $1/\ell|z_i|\approx2\,\widetilde{x}_1^i$ it means that all the terms involving $\widetilde{x}_1^i$ must be replaced by $1/\widetilde{x}_1^i$ in (61a).

The main consequence is the disappearance of the exponential term $\exp\left(-1/2\widetilde{x}_1^i\right)$:

$$u_{i,\ell\pm1} = -\frac{J_i}{|J_i|}\left(1\mp\frac{1}{|z_i|}+f\left(\frac{1}{\widetilde{x}_1^i}\right)\right),\ \text{(QCR)}\ \widetilde{x}_1^i>>1. \quad (61b)$$

We then deal with the *Quantum Critical Regime* (QCR).

When $\widetilde{x}_1^i<1$ we have numerically studied the preponderance of terms $2\,\widetilde{x}_1^i$ and $\frac{8\pi}{e}\left(1-\widetilde{x}_1^i\right)\exp\left(-1/2\widetilde{x}_1^i\right)$ appearing in (61a). It is reported in Fig.4. When $\widetilde{x}_1^i<<1$ both terms show the same value. It means that we are approaching the *Néel line* close to 0 K.

$$u_{i,\ell\pm1}\mp\frac{J_i}{|J_i|}\left(\frac{1}{|z_i|}+2\widetilde{x}_1^i\right)\approx\left[1-4\widetilde{x}_1^i+\ldots\right],$$

$$\text{Néel line}\ \widetilde{x}_1^i<<1. \quad (62)$$

When $\widetilde{x}_1^i<1$, we deal with the *Renormalized Classical Regime* (RCR) and the term $\exp\left(-1/2\widetilde{x}_1\right)$ becomes preponderant:

$$u_{i,\ell\pm1}\mp\frac{J_i}{|J_i|}\left(\frac{1}{|z_i|}+2\widetilde{x}_1^i\right)\approx\left[1-\frac{8\pi}{e}\left(1-\widetilde{x}_1^i\right)\exp\left(-1/2\widetilde{x}_1^i\right)+\ldots\right],$$

$$\text{(RCR)}\ \widetilde{x}_1^i<1. \quad (63)$$

It clearly appears on Fig.4 that, when $\widetilde{x}_1^i$ approaches unity, the term $2\,\widetilde{x}_1^i$ again becomes preponderant. We then observe the disappearance of the term $\exp\left(-1/2\widetilde{x}_1^i\right)$ as in the Néel state. We tend towards the *Quantum Critical Regime* (QCR).

From now and for simplifying the physical interpretations we restrict our discussion to the particular case $J=J_1=J_2$ without loss of generality (the index i may be then abandoned for the ratios $u_{i,\ell\pm1}$ and $\widetilde{x}_1^i$). We first consider the spin-spin correlation defined by (25) and (26). In the low-temperature limit, these equations must be considered in the infinite ℓ-limit:

$$\left|\left\langle S^z_{\tilde{0}0} S^z_{\tilde{k}k'} \right\rangle\right| \approx 1-(k+k')\left[2\tilde{x}_1+\frac{8\pi}{e}\left(1-\tilde{x}_1\right)\exp\left(-1/2\tilde{x}_1\right)\right], \quad \text{as } T \to 0 \text{ K } (\ell \to +\infty) \quad (64)$$

Near the critical point T_C=0 K the spin-spin correlation behaves as the power law $r^{-(D-2+\eta)}$ where η is the corresponding critical exponent and where the distance r is equal to k+k'. Here we have D=d+1 where d is the dimension of the crystallographic space (d=2). We immediately derive $\eta = -2$. Finally, due to fact that the corrective terms appearing in (64) are lower than unity, the spin-spin correlation may be written as:

$$\left|\left\langle S^z_{\tilde{0}0} S^z_{\tilde{k}k'} \right\rangle\right| \approx \left(1-X\right)^{k+k'}, \text{ as } T \to 0 \text{ K } (\ell \to +\infty), \quad (65a)$$

$$X \approx 2\tilde{x}_1 \text{ Néel line } \tilde{x}_1 << 1,\ X \approx \frac{1}{|z|} \text{ (QCR) } \tilde{x}_1 > 1, \quad (65b)$$

$$X \approx \frac{8\pi}{e}\left(1-\tilde{x}_1\right)\exp\left(-1/2\tilde{x}_1\right), \text{ (RCR) } \tilde{x}_1 < 1. \quad (65c)$$

As expected we have *two kinds of regimes*: i) a T-decreasing law due to the fact that the ratio $\tilde{x}_1$ is equal to $T/(4\pi\rho^{\mathrm{Ch}})$ in the *Néel state* very close to 0 K ($\tilde{x}_1$<<1) where ρ^{Ch} is the spin stiffness (*cf* (53)) or in the *Quantum Critical Regime* (QCR, $\tilde{x}_1$>1); ii) an exponential decreasing law $\exp(-2\pi\rho^{\mathrm{Ch}}/T)$ in the *Renormalized Classical Regime* (RCR). ρ^{Ch} represents the spin stiffness of the model proposed by Chakravarty *et al.* [2a] and Chubukov *et al.* [2b]. The correspondence with our model is $\rho = 2\rho^{\mathrm{Ch}}$ (i.e., $J=2J^{\mathrm{Ch}}$).

Introducing the general expression obtained for the spin-spin correlation given by (65) in that of correlation length (*cf* (28) and (29)) expressed in the low-temperature limit (i.e., in the infinite ℓ-limit) we may write:

$$\frac{\xi}{2a} \approx \beta|J|, \text{ Néel line } \tilde{x}_1 << 1, \text{ (QCR) } \tilde{x}_1 > 1, \quad (66a)$$

$$\frac{\xi}{2a} \approx \frac{e}{8\pi}\frac{\sqrt{2}}{2}\left(1+\frac{T}{4\pi\rho^{\mathrm{Ch}}}\right)\exp\left(\frac{2\pi\rho^{\mathrm{Ch}}}{T}\right), \quad \text{(RCR) } \tilde{x}_1 < 1. \quad (66b)$$

Near the critical point T_C=0 K, the correlation length ξ behaves as the power law $|T-T_C|^{-\nu}$ where ν is the corresponding critical exponent so that in our case $\xi \sim T^{-\nu}$. It means that:

$$\nu = 1, \text{ Néel line } \tilde{x}_1 << 1, \text{ (QCR) } \tilde{x}_1 > 1, \quad (67a)$$

$$\nu \to +\infty, \text{ (RCR) } \tilde{x}_1 < 1. \quad (67b)$$

These results are in perfect agreement with the corresponding ones obtained by the renormalization group method [2a], [2b]. We conclude this study by another important remark. At very low-temperature the dynamic behavior is characterized by spin waves. The corresponding spin wave velocity c is such as:

$$\hbar c = 2\sqrt{d}\left|J^{\mathrm{Ch}}\right|a \quad (68)$$

where d=2 is the dimension of the crystallographic space. Note that with our conventional writing we must replace $2J^{\mathrm{Ch}}$ by J. Thus (66b) may be rewritten as:

$$\frac{\xi}{2a} \approx \frac{e}{8}\frac{\hbar c}{2\pi\rho^{\mathrm{Ch}}}\left(1+\frac{T}{4\pi\rho^{\mathrm{Ch}}}\right)\exp\left(\frac{2\pi\rho^{\mathrm{Ch}}}{T}\right), \quad \text{(RCR) } \tilde{x}_1 < 1. \quad (69)$$

This result has been also obtained by several authors while using the renormalization group method. It is notably recalled by Elstner *et al.* [7]. As a result *that gives a strong validation to our model which is valid whatever the temperature whereas the expressions derived from the renormalization group are only valid near the critical point* T_C=0 K.

We finish our study by considering the static susceptibility defined by (31)–(34). As G=G' we deal with the susceptibility per atom. In the low-temperature limit, these equations must be considered in the infinite ℓ-limit. We have:

$$\chi \approx \frac{\beta G^2}{12}\left[\left(\frac{1+u_{\ell+1}}{1-u_{\ell+1}}\right)^2+\left(\frac{1+u_{\ell-1}}{1-u_{\ell-1}}\right)^2\right], \quad \text{as } T \to 0 \text{ K } (\ell \to +\infty) \quad (70)$$

The behavior of the susceptibility is mainly conditioned by that of the common denominator $D = \left[(1-u_{\ell+1})(1-u_{\ell-1})\right]^2$:

$$D \approx 4^2, J > 0 \text{ (AF couplings)}, \quad (71a)$$

$$D \approx 4^2\left[2\tilde{x}_1+\frac{8\pi}{e}(1-\tilde{x}_1)\exp\left(-1/2\tilde{x}_1\right)\right]^2, \quad J < 0 \text{ (F couplings)}, \quad (71b)$$

where AF (respectively, F) means antiferromagnetic (respectively, ferromagnetic). It is well-know that, near T_C=0 K, the product χT behaves as $\xi_x\xi_y\mathcal{M}^2$ where $\mathcal{M}$ is the magnetic moment per unit cell. In our case, as we restrict our study to J=J_1=J_2, we have ξ=ξ_x=ξ_y where the correlation length is given by (66a) and (66b).

Thus, for 2*d non-compensated moments* i.e., for ferromagnetic (F) or antiferromagnetic (AF) couplings, the inverse of the denominator D of the susceptibility diverges (*cf* (71b)). As a result, χT diverges as ξ^2:

$$\chi T \approx \left(\beta|J|\right)^2, \text{ Néel line } \tilde{x}_1 << 1, \text{ (QCR) } \tilde{x}_1 > 1, \mathcal{M}(T) \neq 0, \quad J > 0 \text{ (AF couplings)}, J < 0 \text{ (F couplings)}, \quad (72a)$$

$$\chi T \approx \exp\left(\frac{4\pi\rho^{\mathrm{Ch}}}{T}\right), \text{ (RCR) } \tilde{x}_1 < 1, \mathcal{M}(T) \neq 0, \quad J > 0 \text{ (AF couplings)}, J < 0 \text{ (F couplings)}. \quad (72b)$$

These results are in perfect agreement with the corresponding ones obtained by the renormalization group method [2a], [2b]. Near the critical point T_C=0 K, the susceptibility χ behaves as the power law $|T-T_C|^{-\gamma}$ where γ is the corresponding critical exponent so that in our case $\chi\sim T^{-\gamma}$. It means that:

$$\gamma = 3,\ \text{Néel line}\ \tilde{x}_1 << 1,\ (\text{QCR})\ \ \tilde{x}_1 > 1, \quad (73a)$$

$$\gamma \to +\infty,\ (\text{RCR})\ \ \tilde{x}_1 < 1. \quad (73b)$$

In that case too, these results are in perfect agreement with the corresponding ones obtained by the renormalization group method.

IV. Experimental test

In a previous paper [3b], we fitted the experimental susceptibilities measured for powder samples concerning a family of compounds $[\{MnL_2(N_3)_2\}_n]$, with the following ligands L=DENA (diethylnicotinamide), 4acpy (4-acetylpyridine) and minc (methylisonicotinate). These compounds are characterized by $2d$ square lattices of classical spins (the manganese ions) isotropically coupled (i.e., showing Heisenberg couplings). These fits have been achieved through the theoretical closed-form expression of the susceptibility given by (31) and restricted to the first term of the ℓ-expansion i.e., ℓ=0. Indeed, in a previous article (see Table I in [4b]), we already compared the reduced Néel température $k_BT_N/\sqrt{|J_1J_2|}$ to 0.255 which is the numerical criterion recalled in Fig.1, for compounds **1** (L=DENA), **2** (L=4acpy) and **3** (L=minc). We concluded that, for the 3 compounds, we always have $k_BT_N/\sqrt{|J_1J_2|} > 0.255$. In this article we have chosen to focus on $[\{Mn(DENA)_2(N_3)_2\}_n]$ because the corresponding Néel temperature T_N=2.0 K is very low. That means that the $2d$ magnetic behavior occurs in the low-temperature region. The corresponding fit of the product χT is reported Fig.5. $[\{Mn(DENA)_2(N_3)_2\}_n]$ is characterized by antiferromagnetic couplings (i.e., compensated magnetic moments) showing a single exchange energy J/k_B=4.15 K (the reduced Néel temperature is 0.482).

Near T_C=0 K, if we use the low-temperature expansion of the ℓ-polynomial giving the susceptibility restricted to ℓ=0 (*cf* (31)-(34)), the product χT may be written as:

$$\chi T \approx \frac{G^2}{3}\left(\frac{k_BT}{2J}\right)^2,\ \text{as}\ T\to 0\ \text{K}. \quad (74)$$

Of course this results proves that we are in the *Quantum Critical Regime* (QCR, $\tilde{x}_1$>1). In that case, Chubukov *et al.* [2b] have obtained:

$$\chi T \approx \frac{G^2}{3(\hbar c)^2}\left(\frac{k_BT}{2}\right)^2\ \text{as}\ T\to 0\ \text{K} \quad (75)$$

where $\hbar c = 2\sqrt{2}\left|J^{\mathrm{Ch}}\right|a$, with $2J^{\mathrm{Ch}}=J$ in our model. It is equivalent to consider a unit cell of spacing $a\sqrt{2}$ and the corresponding surface $(a\sqrt{2})^2$ (which represents the lattice of factors G or G'). Thus $\chi T(a\sqrt{2})^2$ given by (75) and (74) exactly coincide. This is a further strong validation of our theoretical model which is confirmed by an experimental test.

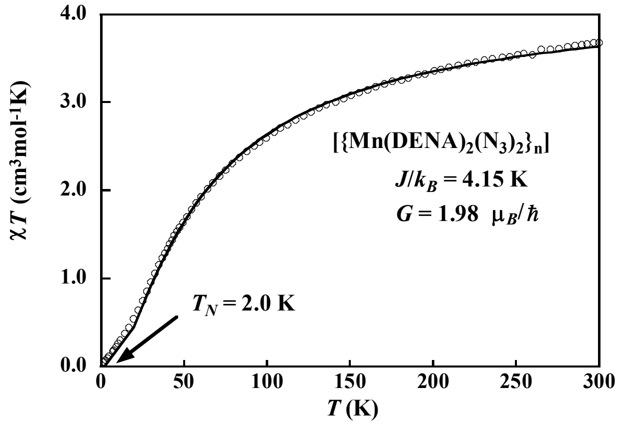

Fig. 5. Theoretical fit of the experimental susceptibility for a powder sample of the compound $[\{Mn(DENA)_2(N_3)_2\}_n]$ characterized by a $2d$ square lattice composed of classical spins (the manganese ions) isotropically coupled (i.e., showing Heisenberg couplings); the ligand DENA is the group diethylnicotinamide.

References

[1] J.G. Bednorz, and K.A. Müller, "Possible high T_C superconductivity in the Ba-La-Cu-O systems," *Z. Phys.* B64, pp. 189-193, 1986.

[2] (a) S. Chakravarty, B.I. Halperin, and D.R. Nelson, "Two-dimensional quantum Heisenberg antiferromagnet at low-temperatures," *Phys. Rev.* B39, pp. 2344-2371, No.4, February 1989; (b) A.V. Chubukov, S. Sachdev, and J. Ye, "Theory of two-dimensional quantum Heisenberg antiferromagnets with a nearly critical ground state," *Phys. Rev.* B49, pp. 11919-11961, No.17, May 1994.

[3] (a) J. Curély, and J. Rouch, "Thermodynamics of the 2*d* Heisenberg classical square lattice. III. Study of the static susceptibility," *Physica*, vol. B254, pp. 298-321, November 1998; (b) J. Curély, "Study of the magnetic properties of a new family of two-dimensional (2D) classical Heisenberg square antiferromagnets," *Proceedings of Optim 2004*, vol. 1, pp. 3-8 (IEEE), Ed. The Transilvania University of Braşov, Faculty of Electrical Engineering and Computer Science, Braşov, Romania, May 2004.

[4] (a) J. Curély, "Study of the magnetic properties of two-dimensional (2*d*) classical square Heisenberg antiferromagnets. I-Zero-field partition function," *Proceedings of Optim 2010*, vol. 1, pp. 66-75 (IEEE), Ed. The Transilvania University of Braşov, Faculty of Electrical Engineering and Computer Science, Braşov, Romania, May 2010; (b) J. Curély, "Study of the magnetic properties of two-dimensional (2D) classical square Heisenberg antiferromagnets. II-Spin correlations and susceptibility," *Proceedings of Optim 2010*, vol. 1, pp. 76-85 (IEEE), Ed. The Transilvania University of Braşov, Faculty of Electrical Engineering and Computer Science, Braşov, Romania, May 2010.

[5] D.A. Varshalovich, A.N. Moskalev, and V.K. Khersonskii, *Quantum Theory of Angular Momentum*, Singapore: World Scientific, 1988.

[6] (a) F.W.J. Olver, The asymptotic expansion of Bessel functions of large order, *Philo. Trans. Roy. Soc. London*, Ser. A247, pp. 328-368 (1954); (b) *Handbook of mathematical functions*, M Abramowitz and I.A. Stegun, Eds. New York: Dover Publications Inc., 9th edition, 1970.

[7] N. Elstner, A. Sokol, R.R.P. Singh, M. Greven, and R.J. Birgeneau, *Spin Dependence of Correlations in Two-dimensional Square-Lattice Quantum Heisenberg Antiferromagnets*, *Phys. Rev. Lett.* 75, pp. 938-941, No 5, July 1995.