Condensed Matter Physics, 2003, Vol. 6, No. 3(35), pp. 551–560

Replica Ornstein-Zernike equations for positionally frozen Heisenberg systems

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Received March 28, 2003

We present the formulation of the Replica Ornstein-Zernike equations for a model of positionally frozen disordered Heisenberg spin system. The results are obtained for various models, one in which the particle positions correspond to a frozen hard sphere fluid, another system in which the configurations are generated by a random insertion of hard spheres, a system of randomly distributed spins, and finally a system corresponding to a soft sphere fluid quenched at high and low temperatures. We will see that the orientational structure of the spin system is fairly well reproduced by the integral equation which, however, does not correctly account for the critical behaviour.

Key words: *Heisenberg system, integral equations, ferromagnetic transition, spinglass*

PACS: *75.50.Lk, 61.20.Ja, 64.60.Fr*

1. Introduction

The study of positionally frozen dipolar fluids has been the focus of various works in recent years [1–4], using both simulation techniques [1,3], mean field theory [2] and quite recently the Replica Ornstein-Zernike (ROZ) integral equation theory [4]. In related works [5,6], the authors focused on the ferromagnetic transition of a positionally frozen Heisenberg spin system, which is amenable to be treated more accurately using simulation techniques adapted for near critical conditions.

In this paper we will explore the capabilities of the ROZ integral equation treatment to describe the ferromagnetic transition in positionally frozen Heisenberg systems. For this purpose we will solve the ROZ equations in the Hypernetted Chain (HNC) approximation for the models simulated in references [5] and [6], namely, Heisenberg spin systems in which the spin positions are frozen according to the configurations of hard sphere (HS) fluids (model A) [5], a random distribution of spins (model B) [5], and soft spheres (SS) quenched at high and low temperatures (models C and D, respectively) [6]. Additionally, we have considered a system in which the spatial distribution is generated by random insertion of non-overlapping hard spheres in a given volume until the desired density is attained (model E). The critical behavior of this system studied by means of Monte Carlo (MC) simulation and finite size scaling analysis is presented in this paper.

The spin-spin interaction in all the models considered is defined by

$$U(r_{12},\omega_1,\omega_2) = -J(r_{12})(\mathbf{s}_1\cdot\mathbf{s}_2), \tag{1}$$

where exchange coupling is given by

$$J(r) = \epsilon\frac{\sigma}{r}\exp[(\sigma - r)/\sigma],$$

$\mathbf{s}_i$ being the unit vector that describes the orientation of the spin i, and ϵ is a positive constant, favoring ferromagnetic alignment. With this, the reduced temperature will be defined by $T^* = k_{\mathrm{B}}T/\epsilon$. For practical purposes $J(r)$ is truncated at $R = 2.5\sigma$. In the case of the SS model $J(r)$ is shifted at the truncation radius, so that $\lim_{r\to R^-} J(r) = \lim_{r\to R^+} J(r) = 0$. As mentioned before, the spatial distribution of model A is simply generated by taking independent HS configurations (in which the particle diameters are $\sigma_0 = \sigma$) at the desired density, which in this case will be $\rho\sigma^3 = 0.6$. The random positions of model B are generated by simple insertion of hard spheres of diameter $\sigma_0 = 0.1\sigma$ in a given volume until the density $\rho\sigma^3 = 0.6$ is reached. These configurations are practically random and the small hard cores prevent the divergence of the interactions at zero separation. Models C and D are generated by placing the spins onto the positions of a frozen SS fluid which interacts via a potential of the form

$$\psi_{\mathrm{soft}}(r) = 4\epsilon\left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right] + \epsilon, \qquad r < 2^{1/6}\sigma$$

and $\psi_{\mathrm{soft}}(r_{12}) = 0$ otherwise. Now, in this case the spatial distribution of the spins will be defined by quenching the positions of the SS fluid at a quench temperature $T_0^* = kT_0/\epsilon$ (with $T_0^* = 2.1$ for model C and $T_0^* = 100$ for model D). As a matter of fact, in the case of Heisenberg interactions, the spatial distribution is hardly sensitive to the spin-spin correlations for temperatures as low as the Curie point. One might envisage then the situation depicted here as the effect of quenching the particle positions on the spin fluid itself, and the same can be said for the frozen HS Heisenberg system. Finally, model E is constructed exactly as the random system B, but using a particle diameter $\sigma_0 = \sigma$. The distribution thus generated has some similarities with the HS model but is somewhat more random.

The rest of the paper is organized as follows. In the next section we introduce the ROZ equations for the positionally frozen Heisenberg system. In section 3 we present our most significant results and conclusions.

2. A summary of the theory

The application of the replica trick [7] to a system like ours, in which the orientational degrees are allowed to equilibrate and the particle positions are frozen, implies taking the $s \to 0$ limit of a fully equilibrated system composed of soft spheres with embedded s replicas of the spins. The Hamiltonian of such a system reads

$$H^{\text{rep}} = \frac{\beta_0}{\beta} \sum_{i>j} \psi_0(r_{ij}) + \sum_{\alpha=1}^{s} \sum_{i>j} J(r_{ij})(\mathbf{s}_i^{\alpha} \cdot \mathbf{s}_j^{\alpha}), \tag{2}$$

where $\beta = 1/k_{\text{B}}T$ and $\beta_0 = 1/k_{\text{B}}T_0$, T_0 and T being the temperature at which the particle positions have been frozen, and the equilibrium temperature of the spins, respectively. In equation (2), $\psi_0(r)$ is either the hard sphere potential, $\psi_{\text{soft}}(r)$, or a given radial potential capable of generating the spatial distributions of models B or E. As prescribed by the replica procedure, only replicas of the same family, α, interact.

The Ornstein-Zernike equation in Fourier space for the replicated system reads

$$\tilde{h}^{\text{rep}}(12) = \tilde{c}^{\text{rep}}(12) + \rho \int \tilde{c}^{\text{rep}}(13)\tilde{h}^{\text{rep}}(32)\text{d}\{\omega_3\}, \tag{3}$$

where $\text{d}\{\omega_3\} = \text{d}\omega_3^1 \ldots \text{d}\omega_3^s$ denotes the integration over the orientations of the s replicas of the spin in particle 3, ρ is the number density, and $\tilde{h}^{\text{rep}}$ and $\tilde{c}^{\text{rep}}$ are the Fourier transforms of the total and direct correlation functions, respectively. The replicated functions can be expanded in Legendre polynomials as

$$f^{\text{rep}}(12) = \sum_{\alpha\beta} \sum_{\text{l}} f_{\text{l}}^{\alpha\beta}(r) P_{\text{l}}(\cos\theta_{12}^{\alpha\beta}), \tag{4}$$

with $f_0^{\alpha\beta}(r) = f_0(r)/s^2$, and in general

$$f_{\text{l}}^{\alpha\beta}(r) = \frac{2l+1}{2} \int \text{d}\cos\theta_{12}^{\alpha\beta} f^{\text{rep}}(12) P_{\text{l}}(\cos\theta_{12}^{\alpha\beta}). \tag{5}$$

Inserting (4) in equation (3) and taking the limit $s \to 0$, the ROZ equations read

$$\begin{aligned}
\tilde{h}_0 &= \tilde{c}_0 + \rho\tilde{c}_0\tilde{h}_0\,, & (6)\\
\tilde{h}_{\text{l}}^f &= \tilde{c}_{\text{l}}^f + \frac{\rho}{2l+1}\left[\tilde{c}_{\text{l}}^f\tilde{h}_{\text{l}}^f - \tilde{c}_{\text{l}}^b\tilde{h}_{\text{l}}^b\right], & (7)\\
\tilde{h}_{\text{l}}^b &= \tilde{c}_{\text{l}}^b + \frac{\rho}{2l+1}\left[\tilde{c}_{\text{l}}^f\tilde{h}_{\text{l}}^b + \tilde{c}_{\text{l}}^b\tilde{h}_{\text{l}}^f - 2\tilde{c}_{\text{l}}^f\tilde{h}_{\text{l}}^f\right], & (8)
\end{aligned}$$

where $l \neq 0$, and

$$\begin{aligned}
f_{\text{l}}^b(r) &= \lim_{s\to 0} f_{\text{l}}^{\alpha\beta}(r), \qquad \alpha \neq \beta,\\
f_{\text{l}}^f(r) &= \lim_{s\to 0} f_{\text{l}}^{\alpha\alpha}(r).
\end{aligned}$$

As usual, the connected functions are defined by $f^c = f^f - f^b$, by which the last two equations transform into

$$\begin{aligned}
\tilde{h}_{\text{l}}^f &= \tilde{c}_{\text{l}}^f + \frac{\rho}{2l+1}[\tilde{c}_{\text{l}}^c\tilde{h}_{\text{l}}^f + \tilde{c}_{\text{l}}^b\tilde{h}_{\text{l}}^c], & (9)\\
\tilde{h}_{\text{l}}^c &= \tilde{c}_{\text{c}}^c + \frac{\rho}{2l+1}\tilde{c}_{\text{l}}^c\tilde{h}_{\text{c}}^c. & (10)
\end{aligned}$$

These equations are just a particular case of those derived by Klapp and Patey [4] for the dipolar case.

As to the closure relation, here we will use the HNC approximation, which reads

$$c^{\mathrm{rep}}(12) = h^{\mathrm{rep}}(12) - \log[g^{\mathrm{rep}}(12)] - \beta u^{\mathrm{rep}}(12), \tag{11}$$

where according to (2)

$$\beta u^{\mathrm{rep}}(12) = \beta_0 \psi_0(r_{12}) + \beta \sum_\alpha J(r_{12})(\mathbf{s}_1^\alpha \cdot \mathbf{s}_2^\alpha). \tag{12}$$

The $s \to 0$ limit of (11) can be obtained if one uses Fries and Patey's form of the HNC closure [8], which expanded in Legendre's polynomials leads to

$$\begin{aligned}
c_0(r) &= -\beta_0 u_0(r) - \int_r^\infty \mathrm{d}r' h_0(r') \frac{\partial X_0(r')}{\partial r'}, & (13)\\
c_l^f(r) &= -\beta u_l(r) - (2l+1) \sum_{\lambda\lambda'} \begin{pmatrix} \lambda & \lambda' & l \\ 0 & 0 & 0 \end{pmatrix}^2 \int_r^\infty \mathrm{d}r' h_\lambda^f(r') \frac{\partial X_{\lambda'}^b(r')}{\partial r'}, & (14)\\
c_l^b(r) &= -(2l+1) \sum_{\lambda\lambda'} \begin{pmatrix} \lambda & \lambda' & l \\ 0 & 0 & 0 \end{pmatrix}^2 \int_r^\infty \mathrm{d}r' h_\lambda^b(r') \frac{\partial X_{\lambda'}^b(r')}{\partial r'} & (15)
\end{aligned}$$

after the limit $s \to 0$ is taken. Above we have used $X(12) = h(12) - c(12) - \beta u(12)$, and the quantities in brackets are the $3-j$ Wigner symbols, $u_0(r) = \psi_0(r)$ and $u_1(r) = J(r)$ (with $u_l = 0$ for $l > 1$.) From equations (15) and (6)–(8) it turns out that $h_l^b = c_l^b = 0$ when $l \neq 0$ (and $h_0^f = h_0^b = h_0$ by definition). With this, the equations can be recast in the form

$$\begin{aligned}
h_0(r) &= \exp[-\beta_0 u_0(r) + s_0(r)] - 1, & (16)\\
h_l^f(r) &= \frac{2l+1}{2} \int \mathrm{d}\cos\theta P_l(\cos\theta)(h_0(r)+1) & \\
&\quad \times \exp\left[\sum_{l'=1} \left(-\beta u_{l'}(r) + s_{l'}^f(r)\right) P_{l'}(\cos\theta)\right], & (17)
\end{aligned}$$

where $s = h - c$, and equation (17) holds for $l > 0$. Equation (6) can be solved coupled with (16) and this gives the spatial distribution of the particles at the quenching inverse temperature β_0. Equations (17) and (7) will describe the orientational structure of the spins at the equilibrium inverse temperature β.

3. Results

The ROZ-HNC equations have been solved by a standard mixing iterates procedure, with the r-space discretized in 8192 points and a grid size of 0.01σ. The spatial distribution of the spins can be visualized in figure 1, where we have plotted the pair distribution function, $g_0(r) = h_0(r) + 1$ for models A–E. The only features worth mentioning are, on the one hand, the slight difference between the $g_0(r)$ of models A and E, despite the fact that the hard sphere configurations are generated by

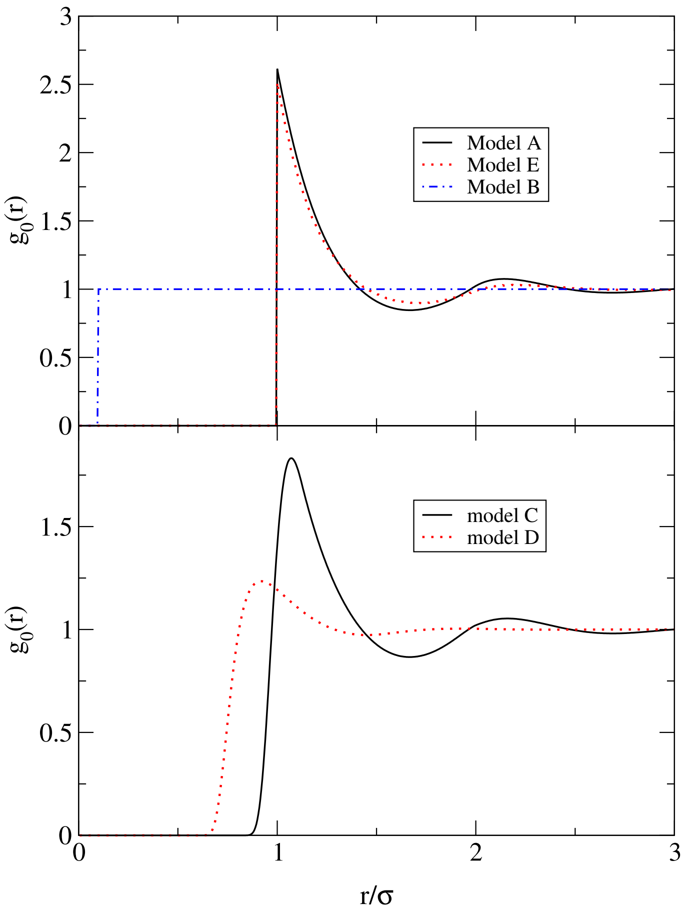

Figure 1. Radial distribution function of the positionally frozen Heisenberg systems studied in this work.

completely different procedures (this simply illustrates how the hard core condition completely dominates the structure of the fluid even at relatively low densities). On the other hand, the comparison between the high and low temperature SS quench shows that the high temperature quench allows for a closer approach distance between the particles, which will have consequences on the critical temperature.

The results of the ROZ equations for the structure of the positionally frozen Heisenberg system are illustrated in figure 2 where the most significant angular coefficient of the fluid-fluid correlation function, $h_1^f(r)$, is depicted. This coefficient is proportional to the ensemble average $\langle \mathbf{s}_1 \cdot \mathbf{s_2} \rangle(r)$ which measures the relative orientation of the two spins separated by a distance r. The results presented correspond to the SS model D, in which the spatial correlations of the quenched system differ most notably from those of the corresponding fully equilibrated (i.e. spin fluid) system. In contrast, the correlations for the HS quenched systems are completely dominated by the HS repulsion (which is temperature independent) and thus the structures of quenched and equilibrated systems are extremely similar. We observe that the ROZ equation rather accurately reproduces the structure for the highest temperature. As

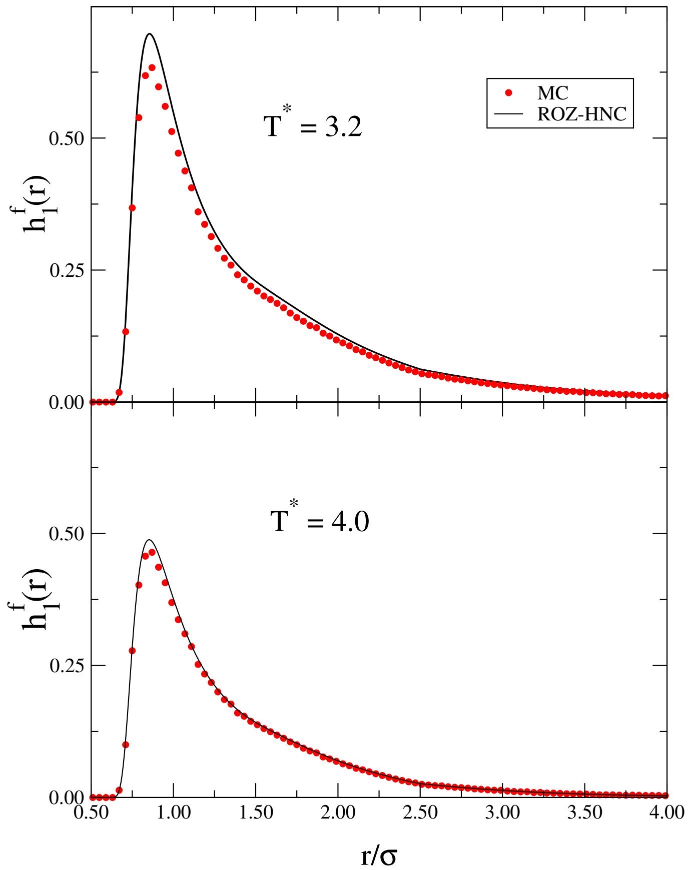

Figure 2. Leading angular coefficient of the pair distribution function expansion for the positionally frozen Heisenberg system constituted by spins embedded in frozen soft spheres (high temperature quench, model D).

the temperature is lowered the theoretical correlation becomes somewhat more long ranged that its MC counterpart. We will see that this is consistent with a systematic overestimation of the critical temperature on the part of the ROZ results.

The critical parameters of model E have been calculated following the prescription of references [5] and [6] (essentially similar to the procedure used in the spin fluid cases studied in reference [10] and [9]). In figure 3 we present the evolution with temperature of Binder's cumulant [11], U_4, and the percolation fraction, ϕ, (defined as the fraction of configurations for which, at least one of the Swendsen-Wang clusters percolates through the periodic system) for various system sizes. Binder's cumulant is defined by

$$U_4 = 1 - \frac{\langle m^4 \rangle}{3\langle m^2 \rangle^2}, \tag{18}$$

where the magnetization per particle is

$$m = \frac{1}{N}\Big|\sum_j^N \mathbf{s}_i\Big|,$$

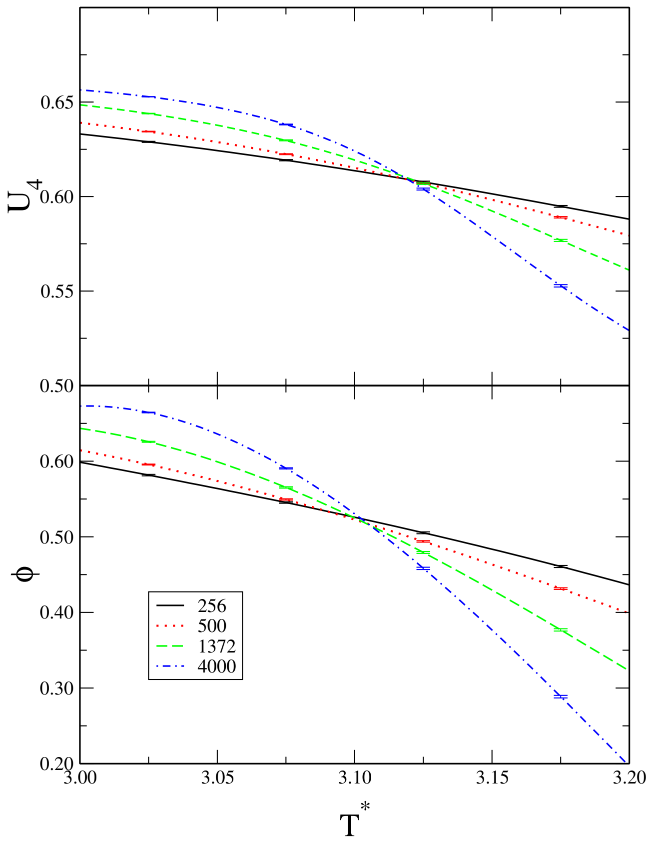

Figure 3. Evolution of Binder's cumulant, U_4, and percolation fraction,ϕ, with temperature and system size for the positionally frozen Heisenberg system with the spatial distribution described by model E (randomly inserted non-overlapping hard spheres of diameter $\sigma_0 = \sigma$).

N being the number of particles in the sample. From the analysis of the intersections of the U_4 and ϕ curves one obtains the critical parameters for this model which are presented in table 1 together with the parameters for the rest of the models taken from references [5] and [6], as well as the results for the hard sphere Heisenberg spin fluid from reference [9], and for the soft sphere spin fluid of reference [10]. The values of β/ν are obtained by a fit of the critical magnetization, m_{c}, which is known to scale with the system size as $m_{\mathrm{c}} \propto L^{-\beta/\nu}$, and γ/ν is obtained from a fit of the magnetic susceptibility maxima near the critical point, which also scale with system size as $\chi_m \propto L^{\gamma/\nu}$. In table 1 we also include the values of T^*_{ns}, the temperature at which the ROZ integral equation breaks down with diverging zero-field magnetic susceptibility,

$$\chi/\rho\beta\mu^2 = \frac{1}{3}\left[1 + \frac{\rho}{3}\tilde{h}^f_1(0)\right] = \frac{1}{3}\left[1 - \frac{\rho}{3}\tilde{c}^f_1(0)\right]^{-1}, \tag{19}$$

where μ is the coupling parameter between the spins and the external field. The first striking feature in table 1 is the fact that despite the equilibrium HNC approximation predicts the transition temperatures within less than a 6% deviation from the

Table 1. Critical parameters of positionally frozen Heisenberg systems obtained from FSS analysis of the MC simulation, and non-solution temperature of the ROZ-HNC (or HNC for the spin fluid systems) equations, $T^*_{\rm ns}$.

Model	T^*_0	σ_0/σ	$T^*_{\rm c}$	U_4	γ/ν	β/ν	$T^*_{\rm ns}$
A [5]	–	1.0	3.145(3)	0.609(3)	1.87(2)	0.53(1)	3.517
B [5]	–	0.1	3.947(2)	0.616(4)	1.86(1)	0.55(2)	6.520
E	–	1.0	3.120(3)	0.609(2)	1.86(2)	0.57(1)	3.463
HS spin fluid [9]	–	1.00	3.150(5)	0.608(1)	1.85(1)	0.56(2)	3.186
C [6]	2.1	–	2.050(6)	0.617(2)	1.92(2)	0.52(4)	2.420
D [6]	100.0	–	2.196(3)	0.618(2)	1.93(2)	0.51(4)	2.826
SS spin fluid [10]	–	–	2.054(1)	0.619(2)	1.90(3)	0.54(2)	2.187

simulation in the spin fluid cases, the theoretical results for the quenched systems are considerably away from the simulated estimates. As a general trend the ROZ overestimates the critical temperatures, but the results are particularly off in the case of the random model B. Moreover, comparing model A with the HS spin fluid, and model C with the corresponding SS spin fluid, one observes that the ROZ predicts an increase in the critical temperature when quenching the particle positions for these systems with positional correlations. This is in agreement with the ROZ results reported in reference [4] for dipolar systems, but it is in clear disagreement with the simulation results that unequivocally indicate that freezing the particle positions does not affect the critical behaviour, at least in the case of separable angular interactions. Aside from this, what seems to be correct in the ROZ predictions is the qualitative change in the critical temperature when comparing different positionally frozen systems. Thus, we observe that the ROZ critical temperatures are ordered as $T^*_{\rm ns}(C) < T^*_{\rm ns}(D) < T^*_{\rm ns}(E) < T^*_{\rm ns}(A) < T^*_{\rm ns}(B)$. This is the same order observed in the simulation results. Moreover, when comparing models E and A, one observes that $T^*_{\rm ns}(E) < T^*_{\rm ns}(A)$. The only difference between these two models lies in the fact that model A presents fluid-like positional correlations, whereas model E is somewhat more random (hardly noticeable in the distribution functions, cf. figure 1). This agrees with the results reported by Klapp and Patey for the dipolar fluid [4] who based on their ROZ calculations concluded that the presence of positional correlations tends to increase the critical temperature. Interestingly, the simulation results also support this view. As a general observation, it can be mentioned that the marked increase in $T^*_{\rm c}$ when going from model C to D and, very especially, from models A or E to model B, simply reflects the fact that the core size in models D and B is smaller (cf. figure 1), by which particles can get in closer contact, thus enhancing angular correlations and subsequently rising the critical temperature.

In summary, we have presented the results of the ROZ-HNC integral equation for a variety of positionally frozen Heisenberg systems in order to assess its ability to describe the critical behaviour. Although the theory yields reasonably accurate results for the structure, it presents a severe deficiency: it systematically predicts a substantial increase in the critical temperature due to the freezing of the particle positions, in contrast to the evidence provided by the simulation. As a consequence, all the theoretical critical temperatures are overestimated by far. On the other hand, when considering just the positionally frozen systems, the ROZ theory predicts the correct dependence of the critical temperature on the topology. Finally, both theory and simulation seem to support the view that positional correlations tend to raise the critical temperature.

4. Acknowledgement

Dr. N.G.Almarza is gratefully acknowledged for his assistance in the simulation calculations. The authors acknowledge support from the Dirección General de Investigación Científica y Técnica under grant No. BFM2001-1017-C03-01.

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Реплічні рівняння Орнштейна-Церніке для позиційно заморожених гайзенбергівських систем

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Отримано 28 березня 2003 р.

Ми формулюємо реплічні рівняння Орнштейна-Церніке для моделі з позиційно замороженою невпорядкованою гайзенбергівською спіновою системою. Результати отримані для різних моделей, в одній з яких положення частинки відповідає замороженому твердосферному плину. Інша система, в якій конфігурація генерується випадковим вміщенням твердих сфер, також система з випадково розподіленими спінами і нарешті система, яка відповідає плину м'яких кульок, розглянуті при високих і низьких температурах. Ми можемо бачити, що орієнтаційна структура спінової системи досить добре відтворюється методом інтегральних рівнянь, які, проте, не дають коректного опису критичної поведінки.

Ключові слова: *гайзенбергівська система, інтегральні рівняння, феромагнетний перехід*

PACS: *75.50.Lk, 61.20.Ja, 64.60.Fr*