A SPIN-ONE LATTICE MODEL OF MICROEMULSIONS AT LOW TEMPERATURES

Roman Kotecký, Lahoussine Laanait,
Alain Messager, Salvador Miracle-Solé

Charles University, Praha, Ecole Normale Supérieure, Rabat,
Centre de Physique Théorique CNRS, Marseille

Abstract: The phase diagram of a three state microemulsion model at low temperatures is discussed. It is shown how, taking into account low energy excitations, the ground state phase diagram is modified and the degeneracy of a coexistence line bordering the region of lamellar phase is taken away.

Key Words: microemulsion, Gompper-Schick model.
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1. Introduction

Three component mixtures of water, oil and amphiphiles exhibit very interesting behaviour as temperature and concentration of the surfactant are varied. For a microscopic theory of these systems several hamiltonians have been proposed [1-6]. In a first model, proposed by Widom [1, 2], formulated in terms of Ising variables, the three species of molecules occupy, with some constraints, the bonds of a lattice. More recently, a three component lattice model has been proposed by Schick, Shih and Gompper [3, 4] in order to illuminate some additional aspects of amphiphilic systems, among which are the microemulsion phase and the interfacial properties.

Previous studies [3, 4] based on a mean field theory present an essential first step towards understanding the behavior of the model. However, some features, as the infinite degeneracy of the zero temperature state manifold occurring for some values of the amphiphile strength have not been covered by the mean field approach.

In this note we wish to show how one can account for fluctuations beyond the mean field and to study rigorously, whenever it is possible, the phase diagram of the model. Namely, we use the standard low temperature perturbation [7] in order to understand the influence of local excitations and to study the phase diagram.

The considered model is a simple three component lattice system in dimension $d \geq 3$. To each site of a cubic lattice $\mathbb{Z}^d$ is assigned a spin one variable s_i so that the values $s_i = 1, -1, 0$ correspond to the presence at site i of a molecule of water, oil, or amphiphile, respectively. The hamiltonian is

$$\mathcal{H} = \sum_{(i,j)} J(s_i - s_j)^2 + \sum_i (Bs_i^2 - Hs_i) + \sum_{(i,j,k)} Ls_i(1 - s_j^2)s_k.$$

The first term is the sum of pair interactions, assumed to be attractive between particles of the same kind ($J > 0$), and the second one contains the usual chemical potentials. With these two terms the hamiltonian is a particular case of the Blume-Emery-Griffiths model [8], which describes a simple three component mixture for the case of nearest neighbour interactions. Even though we assume that two particles of different kind have the same pair interactions, the discussion for general pair interactions can be done by following the same method which is used in the present work. The external field H is related to the chemical potential difference between oil and water and B is related to the chemical potential of the surfactant. The third sum extends over the sets (i, j, k) of three adjacent sites in a line and $L > 0$ is the strength of the amphiphilic interaction. This term distinguishes the molecule associated to spin 0 as an amphiphile and mimics its effect by favouring the configuration with 0 placed between $+$ and $-$, all in a line.

2. Phase diagram with low energy excitations

The *ground states* of the model are described as follows. If we assume that $L = 0$, the spins must be equal everywhere since $J > 0$. The system has three ground states, the (0), (+) and (−) states, where respectively s_i is equal to 0, +1 and −1 for all i. The energies per site in these states are

$$h^{(0)} = 0 \quad , \quad h^{(+)} = B - H \quad , \quad h^{(-)} = B + H$$

and the corresponding phase diagram is easily obtained.

The amphiphile interaction favorizes the configuration $(+0-)$ on three aligned adjacent sites (i, j, k). Such configurations appear in ground states if

$$L - 2J > 0$$

and, for this reason, we assume hereafter that this condition is satisfied. Then, at least for some values of B and H, it is clear that the system would favour to have the L-bonds (or the sets of three adjacent sites) occupied as much as possible by such $(+0-)$ configurations. This leads to a new family of ground states with a lamellar structure. To describe them more precisely we introduce some notations. We consider the planes $r(i) = \alpha_1 i_1 + \ldots + \alpha_d i_d = z$, where all α's are +1 or −1 and z is an integer. These planes will be called *diagonal planes*, they may have 2^{d-1} possible orientations. We consider the sequence of all diagonal planes of the lattice with a given fixed orientation. The configurations that assume a constant value in each plane of such a sequence will be called *layered* configurations. The *lamellar* ground states are the layered configurations obtained by putting all spins equal to 0 on every second plane of the sequence and alternatively equal to +1 and to −1 on the remaining planes. In other words, the sequence of spin values on the diagonal planes is the periodic sequence $(0+0-\ldots)$ of period 4. This leads to $4.2^{d-1} = 2^{d+1}$ equivalent (i.e., related by translations and rotations of the lattice) lamellar ground states. Their energy per site is

$$h^{(\mathrm{lam})} = -\frac{d}{2}(L - 2J) + \frac{1}{2}B.$$

By comparing this expression with those corresponding to the other ground states we obtain the following grond state phase diagram (see Fig. 1).

The lamellar states are the unique ground states, for values of B and H in the triangular region

$$\frac{B}{d} < L - 2J \qquad \frac{2H}{d} < L - 2J + \frac{B}{d} \qquad -\frac{2H}{d} > L - 2J + \frac{B}{d}$$

Outside this region we have the $(+)$ state, if $H \geq 0$ and $H \geq B$, the $(-)$ state, if $H \leq 0$ and $H \leq B$ and the (0) state, if $|H| \leq B$.

We next examine the coexistence lines of the phase diagram. On the separation line between the (0) and (lam) states, only these two states are ground states. The same situation occurs on the coexistence lines $(0)/(+)$, $(0)/(-)$ and $(+)/(-)$. However, on the coexistence lines $(+)/(\mathrm{lam})$ and $(-)/(\mathrm{lam})$ infinitely many ground states occur. All of them have a layered structure and may be described by the sequence of spin values associated to the sequence of parallel diagonal planes. A particular role will be played by the periodic ground states associated to the periodic sequences $(0 + \ldots + 0 - \ldots -)$, in which $m \geq 1$ diagonal planes, where the spins are $+$, are separated from $n \geq 1$ diagonal planes, where the spins are $-$, by single planes, where the spins are 0. We use the notation (m, n) for these states of period $p = m+n+2$. The periodic state (1,1) is the (lam) ground state considered above. The energies per site of these states are

$$h^{(m,n)} = h^{(+)} - \frac{2d}{p}\,(L - 2J) - \frac{2}{p}\,B + \frac{2(n+1)}{p}\,H.$$

This shows that all ground states with $(m, n = 1)$, and $(m = 1, n)$, are present, respectively, on the coexistence lines $(+)/(\mathrm{lam})$ and $(-)/(\mathrm{lam})$. All these states, for any m and n, are present at the triple point, where these two lines and the $(+)/(-)$ coexistence line intersect.

Our aim is to show that the considered model can be, in the region $L - 2J > 0$, rigorously analyzed at low temperatures. The phase diagram at low temperatures follows from a competition between energies of ground states taking into account additional contributions of entropies of low energy excitations. These ideas are formalized in the Pirogov-Sinai theory of low temperature phase diagram [7]. Here we will use an extension of this theory due to Bricmont and Slawny [9].

This extension concerns a mechanism of supression of high energy fluctuations. We first introduce some notions needed for the study of equilibrium states at low temperatures. By an *excitation* we understand a configuration which coincides with a ground state outside of a finite set of sites. If, in a given system, the energy of an excitation (relative to the corresponding ground state) tends to infinity with the size of the region where it differs from the surrounding ground state, this system is said to be *regular**. The Peierls condition, required in the Pirogov-Sinaï theory is satisfied in a given system, if the energy of an excitation is proportional to the size of the boundary of this region (or larger). Clearly, all models in which

* Notice, however, that an excitation may consists of a region where the configuration is a ground state, but different from that surrounding the region from outside.

the Peierls condition holds are regular. The method of Bricmont and Slawny applies to a more general class of regular systems than those satisfying the Peierls condition. We shall show that this is the case for the model under consideration which is regular but has an infinite number of ground states and fails to satisfy the Peierls condition. The two dimensional version of the model is not even regular, the lack of regularity which occurs also in the one-dimensional Ising model.

It is useful to consider the excitations as partial or local configurations on the lattice, consisting of the set of excited sites, i.e. those which contribute to the increase of the energy with respect to that of the ground state, and an appropriate boundary around this set. The precise definitions are as follows. We define a *partial configuration* X by specifying a finite set of sites, called the *domain* of X, $\mathrm{dom}(X)$, together with a configuration on this set. We define a subset of $\mathrm{dom}(X)$ called the *boundary* of X, $\partial(X)$, in such a way that if the configuration on $\partial(X)$ is kept fixed, the sites in $\mathrm{dom}(X) \setminus \partial(X)$ do not interact with the sites outside of $\mathrm{dom}(X)$. A standard way is to define $\partial(X)$ as an ℓ-boundary, with $\ell \geq 2$, i.e., as the set of sites in $\mathrm{dom}(X)$ at distance less than ℓ from its complementary set. A partial configuration X is an *excitation* if its restriction to $\partial(X)$ is a ground configuration on $\partial(X)$.

We next present some simple examples assuming only one excited site in the ground state. We add to this excited site a boundary consisting of its nearest neighbours and also the next nearest neighbours on the same line when the excited site interacts (by an L-bond) with them. We get, for instance the following partial configurations (actually there are, up to translations and rotations of the lattice, fourteen possibilities for such one site excitations).

$$
\begin{array}{cccc}
\begin{array}{cccc} & & & \\ & & + & \\ - & 0 & 0 & + \\ & & 0 & \\ & & - & \end{array}
&
\begin{array}{cccc} & & & \\ & & - & \\ + & 0 & 0 & - \\ & & 0 & \\ & & + & \end{array}
&
\begin{array}{ccccc} & & - & & \\ & & 0 & & \\ - & 0 & 0 & 0 & - \\ & & 0 & & \\ & & - & & \end{array}
&
\begin{array}{ccccc} & & + & & \\ & & 0 & & \\ + & 0 & 0 & 0 & + \\ & & 0 & & \\ & & + & & \end{array}
\\
X_1 & X_2 & X_3 & X_4
\end{array}
$$

Although only two dimensions are shown here, our intention is to represent d-dimensional objects that can easily be understood from the figures, taking into account the layered structure of ground configurations.

We observe that all these excitations have the property of being removable: an excitation X is *removable* if there exists a unique ground configuration on dom(X) denoted $G(X)$, whose restriction to $\partial(X)$ is equal to the restriction of X. If X is a removable excitation, then its energy $E(X)$, relative to the ground state, is well defined. It is the energy of the configuration X minus the energy of $G(X)$. If X is a removable excitation and Y a partial configuration whose domain contains dom(X) and such that its restriction to dom(X) is equal to X, then there is a unique partial configuration obtained by removing X from Y, equal to $G(X)$ on dom(X) and to Y otherwise. The ground configurations $G_k = G(X_k)$, $k = 1, ..., 4$, associated to the excitations considered above, are the following ones

$$
\begin{array}{cccc}
 & & & \\
 & & + & \\
- & 0 & + & + \\
 & & 0 & \\
 & & - & \\
 & & & \\
 & & G_1 &
\end{array}
\qquad
\begin{array}{cccc}
 & & & \\
 & & - & \\
+ & 0 & - & - \\
 & & 0 & \\
 & & + & \\
 & & & \\
 & & G_2 &
\end{array}
\qquad
\begin{array}{ccccc}
 & & - & & \\
 & & 0 & & \\
- & 0 & + & 0 & - \\
 & & 0 & & \\
 & & - & & \\
 & & & & \\
 & & G_3 & &
\end{array}
\qquad
\begin{array}{ccccc}
 & & + & & \\
 & & 0 & & \\
+ & 0 & - & 0 & + \\
 & & 0 & & \\
 & & + & & \\
 & & & & \\
 & & G_4 & &
\end{array}
$$

and the energies $E_k = E(X_k)$, $k = 1, ..., 4$, of the excitations are

$$
\begin{array}{ll}
E_1 = -B + H + dL, & E_2 = -B - H + dL, \\
E_3 = -B + H + 2dL - 2dJ, & E_4 = -B - H + 2dL - 2dJ.
\end{array}
$$

Since $dL - 2dJ > 0$, we have $E_1 < E_3$ and $E_2 < E_4$. Moreover, when we consider the system in the vicinity of the $(+)/(\mathrm{lam})$ or the $(-)/(\mathrm{lam})$ coexistence lines of the phase diagram of the ground states, only the elementary excitations X_1,...,X_4, have energy less or equal than $E = \max\{E_3, E_4\}$. This occurs at least for some range of values of the coupling constants (namely, if $L < (8/3)J$) to which, for concreteness, we shall restrict our discussion.

If X is a configuration of the system equal to a ground configuration G outside Λ, where Λ is a large box on the lattice, then there is a uniquely defined configuration called the *retouch of* X, $\mathrm{ret}(X)$, obtained from X by removing all elementary excitations with energy smaller than a given value E_0 (i.e., in the case considered above, $E_0 = E$, all excitations of types X_1,...,X_4). Let $\mathcal{E}(G, E_0)$ be the set of such configurations X which, moreover, satisfy the condition $\mathrm{ret}(X) = G$. Namely, it is the set consisting of the ground configuration G and all its excitations whose energy locally does not exceed E_0. We assign to every configuration in $\mathcal{E}(G, E_0)$ the corresponding Boltzmann weight and supress the remaining ones by assigning them the zero probability. In this way we obtain a state of the system (in the box Λ), which will be called the *restricted ensemble* associated to the ground state G (with excitations of energy less than E_0). The partition function Z_Λ^{G,E_0}, restricted to the configurations of the set $\mathcal{E}(G, E_0)$, yields the free energy per site associated to this restricted ensemble $f_\Lambda^{G,E_0} = -(1/\beta|\Lambda|) \ln Z_\Lambda^{G,E_0}$ (in this formula $\beta = 1/kT$ denotes the inverse temperature and $|\Lambda|$ is the volume of Λ). This notion extends to the infinite system by taking the limit Λ tending to infinity.

The excitations Y, contributing to a restricted ensemble, have weights $\varphi(Y) = \exp(-\beta E(Y))$. Two excitations are compatible if their domains are disjoint. These facts imply that

$$
Z_\Lambda^{G,E_0} = \exp(-\beta \mathcal{H}(G)) \sum_{\{Y_1,...,Y_k\}} \varphi(Y_1)...\varphi(Y_k)
$$

where $\mathcal{H}(G)$ is the energy of the ground state G and the sum runs over all sets $\{Y_1, ..., Y_k\}$, $(k = 0, 1, 2, ...)$ of compatible excitations of G with energy less then E_0. Therefore, the restricted ensemble can equivalently be described as a polymer system, where the polymers are the different excitations with the activities $\varphi(Y)$.

Since the system is regular and $E(Y) \le E_0$, the number of kinds of these polymers, up to translations, is finite (independent of Λ). The activities $\varphi(Y)$ are small when the temperature is low. We use the convergent small activity expansion (in terms of the Ursell functions) to compute the free energy of the restricted ensemble. Since different ground states may have different excitations, and also different numbers of the common excitations, the free energies of some of the associated restricted ensembles will be different.

Let us consider the case $E_0 = E = \max\{E_3, E_4\}$ in which all the excitations excitations to be taked into account are of types $X_1, ..., X_4$ described above. We use $f^{(m,n),E}$, or simply $f^{(m,n)}$, to denote the corresponding free energy per site when $G = (m,n)$ is one of the periodic ground states of the system. In this case, up to terms of order less than $\exp(-\beta E)$, the low temperature expansion can be limited to the first term for each considered excitation, that is

$$f^{(m,n)} = h^{(m,n)} - \lim_{\Lambda\to\infty} (1/\beta|\Lambda|) \sum_Y \varphi(Y)$$

where $h^{(m,n)}$ is the ground state energy per site and the sum runs over the considered four types of excitations, contained in Λ and contributing to the restricted ensemble $\mathcal{E}(G,E)$ of the ground state $G = (m,n)$. Taking into account the geometric structure of these grounds states, described above, we get

$$\begin{aligned} f^{(m,n)} = {} & h^{(+)} - \frac{2d}{p}\,(L-2J) - \frac{2}{p}\,B + \frac{2(n+1)}{p}\,H - \\ & - \frac{2}{p}\,I(m\ge 2)\varphi_1 - \frac{2}{p}\,I(n\ge 2)\varphi_2 - \frac{1}{p}\,I(m=1)\varphi_3 - \frac{1}{p}\,I(n=1)\varphi_4. \end{aligned}$$

In this formula, the first four terms correspond to the energy per site of the ground state, $\varphi_\ell = (1/\beta)\exp(-\beta E_\ell)$ for $\ell = 1, ..., 4$, and I is the indicator of the condition shown in parentheses (it equals one if the condition is satisfied and is zero otherwise). On the other hand, for G equal to the $(+)$ and $(-)$ states, we have $f^{(+)} = h^{(+)}$ and $f^{(-)} = h^{(-)}$ (there are no excitations of these ground states of energies lower than E).

Having the free energies, we may draw the phase diagram for the restricted ensembles. We say that a point in the (B,H)-plane belongs to the G-state region whenever

$$t^{G,E}(B,H) = f^{G,E}(B,H) - \min_G f^{G,E}(B,H) = 0$$

This phase diagram is schematically represented in Fig. 2.

The region $PQRR'Q'P$ around the point $B = -d(L-2J)$, $H = 0$, is occupied by the restricted ensemble (2,2). For $H \ge 0$ this region is defined by the inequalities

$$\begin{aligned} H - \frac{1}{3}\,(dL - 2dJ + B) &\le \frac{1}{3}\,\varphi_2 + \frac{1}{3}\,\varphi_1, \\ H + \frac{1}{3}\,(dL - 2dJ + B) &\le \frac{5}{3}\,\varphi_2 - \frac{1}{3}\,\varphi_1 - \varphi_4, \\ dL - 2dJ + B &\le 2\varphi_2 + 2\varphi_1 - \frac{3}{2}\,\varphi_4. \end{aligned}$$

The restricted ensemble (2,1) is present in the triangular region QRS, limited by the curve QR already described, and the two curves QS and RS, where

$$-2\varphi_1 + \frac{1}{4}\varphi_4 \leq H - \frac{1}{2}(dL - 2dJ + B) \leq \frac{2}{5}\varphi_1 + \frac{1}{5}\varphi_4.$$

Since the two bounds are inconsistent for low temperatures and $E_4 < E_1$, the region (2,1) ends at a point S, near to the point $B = 0$, $H = (d/2)(L - 2J)$. The curve SW, where

$$H - \frac{1}{2}(dL - 2dJ + B) = \frac{1}{2}\varphi_4,$$

separates, together with the regions above, the (+) and (1,1) restricted ensembles. The lower part ($H < 0$) of the diagram follows by the symmetry with respect to the horizontal axis.

In the phase diagram of Fig. 2, the distance between the nearest curves is of order $\exp(-\beta E)$. If this order were considered to be negligible, we would only observe the region corresponding to the (2,2) restricted ensemble whose size is of order $\exp(-\beta E_2)$. At lower orders the phase diagram reduces to that for the ground states. On the other hand, the restricted ensembles (m, n) associated to the other periodic ground states belong to the coexistence curves PQS and $PQ'S'$ of the diagram of Fig. 2. More precisely, all restricted ensembles with $m \geq 3$ and $n = 1$ coexist on the curve QS, for $m \geq 3$ and $n = 2$ they coexist on the curve PQ, and all states with $m \geq 3$ and $n \geq 3$ coexist at the point P.

3. Conclusions

The discussion presented above can be transformed into a rigorous statement about a full phase diagram (of the model with no restrictions on excitations) at low temperatures. Namely, one can show that there exists β_0, such that for all temperatures $\beta > \beta_0$, there exists, in the plane (H, B), an open region $\Omega(\beta)$ in the complement of the curves PQS and $PQ'S'$ of the phase diagram of the restricted ensembles (Fig. 2), whose distance from these curves is of order less than $\exp(-\beta E)$. In $\Omega(\beta)$ we have a complete phase diagram of the pure thermodynamic phases (extremal periodic Gibbs states of the system) which is a small deformation of the diagram of Fig. 2.

There are in $\Omega(\beta)$, six disjoint open regions, such that their closure jointly covers $\Omega(\beta)$, that correspond to the regions denoted (+),(−) and (m, n) with $m \leq 2$ and $n \leq 2$, in Fig. 2. In each of these regions there is a pure phase which is a small deformation of the associated ground state described above. The boundaries of these regions are smooth open archs in which two distinct phases coexist. They meet, inside $\Omega(\beta)$, in two points (which correspond to the points R and R' of Fig. 2) in which three distinct pure phases coexist. These coexistence curves are deformations of the corresponding curves on Fig. 2 of order $\exp(-\beta E)$.

Moreover, the analysis developped in the present paper can be pursued by considering subsequent excitations of higher energies. This allows to remove degeneracy from the line PQS and obtain a rigorous full description of the phase diagram, in a corresponding region, of the system at low temperatures. This is in fact the main result of our work that we are going to report only briefly in the following paragraphs (a more detailed discussion will be the subject of a separate publication).

First, we prove that, with appropiate definitions of the associated domains and boundaries, and for any E, all elementary excitaions with energy less than E are removable. This allows us to consider restricted ensembles in which all these excitations are taken into account. We may then compute their free energy and, by using the function $t^{G,E}(B,H)$ as explained above, draw the corresponding phase diagram of the restricted ensembles. On the other hand, we prove that, for any given integer k, there is a value $E = E_k$ (which increases linearly with k), such that if all the elementary excitations with energy less than E_k are considered, then a phase diagram may be drawn that distinguishes all the regions belonging to the restricted ensembles associated to the ground states (m,n) for all m and n such that $\max\{m,n\} \leq k$. Then, as in the case of the lowest energy excitations that we have already discussed with some detail, these results lead to a rigorous statement on the equilibrium states of the system at low temperatures. Namely, one can show that there exists $\beta_0 = \beta_0(k)$ (where $\beta_0(k) \to \infty$ when $k \to \infty$) such that for all temperatures $\beta > \beta_0$, there exists in the plane (H,B) an open region $\Omega(\beta)$, in which we have a complete phase diagram of the pure thermodynamic phases, with separated regions for for all pure phases (m,n) for which $\max\{m,n\} \leq k$. This phase diagram is a small deformation of the diagram of the associated restricted ensembles.

A sketch of the phase diagram is shown in Fig. 3. One finds that the phases are ordered according to the increasing values of $(m+n)/p$ when B increases along lines parallel to the B axis while, going along lines parallel to the H axis, the values of $(m-n)/p$ increase when H increases. A first order phase transition takes place when crossing the lines of coexistence which correspond to a discontinuity of one of the order parameters $\langle s_i^2 \rangle$ or $\langle s_i \rangle$. Notice that, as we mentioned above, when k becomes large, the inverse temperature β_0, needed to ensure the existence of the state (m,n) as a separate phase, also becomes larger.

Some similarities between the behaviour of the model considered here and the behaviour of the axial next-nearest-neighbour Ising (ANNNI) model should be clear (see Refs. 10, 11). However, the phase diagram that we were discussing above, is generated in the plane of the surfactant, oil and water chemical potentials, with all interactions fixed. It yields a double infinite sequence of pure phases indexed by the values of m and n. We notice that a rigorous analysis of the model proposed by Widom [1,2] has been recently worked out by Dinaburg and Mazel [12] along lines similar to those developped in our work. They found, in the region that they were able to study, no ANNNI-like behaviour in that model, contrarily to what was surmised in some earlier works (quoted in Ref. 12). In this case, from infinitely many ground states, only a small number of pure phases persists at non-zero temperatures.

The surface tension behaves quite differently in systems with a finite and with an infinite number of ground states. In our case we find that the surface tension goes exponentially to zero as β tends to infinity. This is easy to understand: an interface between two phases does not cost any energy, but it costs the free energy of low-energy excitations. This is, actually, the basic mechanism that justifies the method used in this paper. Since the free energy of these excitations is exponentially small at low temperatures, the same is true for the surface tension.

Finaly, the low surface tension between the oil-rich and water-rich phases, at a given temperature, reflects also the low free energy of the corresponding interface. In fact, since the excitations that distinguish between the ground states (m, n) and $(m-1, n)$ have energy proportional to $k = \max\{m, n\}$, their free energy decays exponentially when k becomes large. Therefore, the surface tension between the phases (m, n) and $(m-1, n)$ is exponentially small for large k.

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Roman Kotecký
Departement of Theoretical Physics, Charles University,
V Holevsovickách 2, 180 00 Praha 8, Czechoslovakia
and
Center for Theoretical Study, Charles University,
Ovocný trh 3, 116 36 Praha 1, Czechoslovakia

Lahoussine Laanait,
Ecole Normale Supérieure,
Takaddoum, Rabat, Marocco

Alain Messager,
Centre de Physique Théorique, CNRS,
Luminy, Case 907, 13288 Marseille Cedex 9, France

Salvador Miracle Solé,
Centre de Physique Théorique, CNRS,
Luminy Case 907, 13288 Marseille Cedex 9, France