Lamellar-like structures in ferrofluids placed in strong magnetic fields

Orion Ciftja\textsuperscript{1,2}

\textsuperscript{1}Department of Physics, Prairie View A&M University, Prairie View, Texas 77446, USA
\textsuperscript{2}Kavli Institute for Theoretical Physics, University of California, Santa Barbara, California 93106, USA

We consider a ferrofluid system consisting of magnetic particles interacting with a magnetic dipole-dipole interaction. We study the strong magnetic field regime where all magnetic dipoles are completely polarized in the direction of the magnetic field. We introduce a lattice gas model that serves to describe space ordering phenomena in such systems. It is found that, within mean field theory, this model predicts a second order phase transition to a phase with inhomogeneous lamellar-like ordering below a certain critical temperature.

PACS numbers: 61.20.Gy, 61.30.Gd, 64.10.+h, 82.70.Dd.
Keywords: A. Magnetically ordered materials; D. Phase transitions.

I. INTRODUCTION

Ferrofluids are stable suspensions of colloidal ferromagnetic particles in suitable, non-magnetic carrier liquids [1, 2]. Each particle consists of a roughly spherical ferromagnetic core coated with a thin layer of inert nonmagnetic material that prevents the system from irreversible aggregation. The dominant interactions are those between the magnetic dipoles [3]. The short-range van der Waals attractions between the coatings are relatively weak, and hence only provide a small sticking effect when two particles come close to contact.

The structure and phase diagram of ferrofluids at zero magnetic fields is essentially determined by the relative strength of: (i) repulsive forces due to coating of particles to prevent aggregation; (ii) attractive van der Waals forces which generally are weak; and (iii) magnetic dipole-dipole interactions which are highly anisotropic and can be attractive/repulsive depending on the relative orientation of magnetic dipoles [4, 5]. Experimental studies [6] have stimulated many theoretical investigations [7–9] and in particular computer simulations have shown that in absence of magnetic field, the magnetic interactions between particles are stronger than a thermal energy, the particles aggregate into chains with magnetic moments of each particle pointing towards a neighbor along the chain [10, 11].

The interaction of ferrofluids with external magnetic fields leads to various interesting and adverse phenomena [12]. Differently from zero or low magnetic field phases of ferrofluids that are mostly homogeneous, the high magnetic field phases are generally inhomogeneous and have pronounced anisotropic properties [13, 14]. An applied magnetic field has a huge impact on ferrofluids and causes them to undergo complicated structural transitions from a randomly dispersed gas to solid structures with either column, bent-wall, or labyrinthine patterns [15]. The precise appearance of these patterns depends on a variety of parameters like interaction strength, applied magnetic field strength, field orientation relative to sample, sample geometry, temperature and volume fraction of emulsion droplets, to mention a few. A systematic understanding of the influence of all these factors on ferrofluid phases is a formidable task both experimentally and theoretically.

An applied magnetic field favors the creation of extended chains oriented parallel to the field direction. In larger magnetic fields, the chains aggregate into thicker columnar structures. Experiments indicate that the distance between columns parallel to the direction of an applied magnetic field decreases for stronger magnetic fields [15]. When the strength of the magnetic field becomes larger than a critical value, labyrinthine or eventually hexagonally ordered structures are observed. In the limit of strong external magnetic fields (of the order of 1 Tesla) all magnetic dipoles are fully aligned in the direction of the magnetic field. In such conditions, dipole-dipole interaction plays the key role and a variety of interesting ordering phenomena can be observed. The stability of these various phases has been analyzed by numerical simulation studies [14, 16] that generally are in qualitative agreement with experiments [17]. However, computer simulations of ferrofluid systems are extremely demanding. Most calculations involve relatively small system sizes that make the observation of regular ordering patterns hard to identify.

Despite the computational shortcomings, there are several numerical simulations [18–21] that show regular pattern formation in the limit of strong magnetic fields. These studies show the appearance of symmetry-breaking structures consisting roughly of planes of particles parallel to the external magnetic field who represent a modulation of the density of such systems (alternating large and small concentration of particles). Thus, to some degree, these structures can be associated with a lamellar phase.

In this work we study a system of ferrofluid particles placed in a strong magnetic field. We introduce a lattice gas model consisting of ferrofluid particles fully aligned in the direction of the applied magnetic field and interacting with an anisotropic dipole-dipole interaction. We develop a mean field theory and show that, below a certain critical temperature, the system has a phase transition from a phase with uniform particle concentration to a phase with alternating larger and smaller concentrations of particles.
that can roughly be related to lamellar ordering. Given that a detailed understanding of phase transitions in ferrofluids is a formidable scientific challenge, such model serves only as a qualitative benchmark.

II. LATTICE GAS MODEL

We consider a system of magnetic particles which are distributed on $N$ sites of a simple cubic lattice with lattice parameters, $\Delta x = \Delta y = \Delta z = a$. The particles are considered to be hard spheres each carrying a central point dipole with magnetic moment, $\mu$. The system is subject to a strong magnetic field. For simplicity we assume that the external magnetic field is applied in the $z$-direction, $\vec{B} = B\hat{e}_z$ where $\hat{e}_x$, $\hat{e}_y$ and $\hat{e}_z$ denote the cartesian unit vectors, respectively, for the $x$, $y$ and $z$ direction. In the strong magnetic field regime (considered throughout this work), all magnetic moments are oriented in the direction of the magnetic field therefore all magnetic moments are $\vec{\mu} = \mu \hat{e}_z$ where $\mu$ is the magnitude of the dipole moment for each particle. The interaction energy between two point dipoles occupying lattice sites $i$ and $j$ is written as:

$$
\epsilon_{ij} = J_{ij} \left[ 1 - 3 \left( \hat{e}_z \cdot \vec{e}_{ij} \right)^2 \right],
$$

where $J_{ij} = \frac{\mu B}{4\pi r_{ij}^3}$ is a positive coupling constant, $\vec{e}_{ij} = \vec{r}_{ij}/r_{ij}$ is a unit vector along the interparticle vector, $\vec{r}_{ij}$ and $r_{ij} = |\vec{r}_{ij}|$ is the distance separating particles $i$ and $j$.

In this study we adopt a lattice gas model where each lattice site, $i$ with coordinate $\vec{r}_i = x_i \hat{e}_x + y_i \hat{e}_y + z_i \hat{e}_z$ has an occupation number, $n(\vec{r}_i)$ which takes the values 1 or 0, depending on whether the lattice site $i$ is occupied or not. This choice mimicks the usual short-range interaction arising from the hard core. Therefore, each lattice site can be either empty or filled with at most one particle. The Hamiltonian of the system can be written as:

$$
H = \sum_{i<j} \epsilon_{ij} n(\vec{r}_i) n(\vec{r}_j) - \mu B \sum_{i=1}^N n(\vec{r}_i).
$$

The first term in the above Hamiltonian represents the sum of all pairs of dipole-dipole interaction energies. The second term is the energy of the aligned dipoles in the direction of the applied magnetic field.

Given that the lattice gas model in Eq.(2) is hardly tractable, we introduce another simplification assuming a dipole-dipole interaction only between particles located at nearest-neighbor sites. Namely, we consider $J_{ij} = J$, if $i$ and $j$ are nearest-neighbor sites and $J_{ij} = 0$ otherwise. With this assumption, we have:

$$
\epsilon_{ij} = \begin{cases} 
J & \text{if } \vec{r}_i - \vec{r}_j = \Delta x \hat{e}_x \\
J & \text{if } \vec{r}_i - \vec{r}_j = \Delta y \hat{e}_y \\
-2J & \text{if } \vec{r}_i - \vec{r}_j = \Delta z \hat{e}_z
\end{cases}
$$

Therefore the Hamiltonian of the system can be written as:

$$
H = \sum_{i=1}^N \left[ J n(\vec{r}_i) n(\vec{r}_i + \Delta x \hat{e}_x) + J n(\vec{r}_i) n(\vec{r}_i + \Delta y \hat{e}_y) -2J n(\vec{r}_i) n(\vec{r}_i + \Delta z \hat{e}_z) \right] - \mu n N B,
$$

where $n = \frac{1}{N} \sum_{i=1}^N n(\vec{r}_i)$ represents the particle concentration. Real ferrofluids are very complicated systems, therefore it should be noted that any theoretical model has to involve several approximations. For instance, a key approximation of the lattice gas model under consideration is to assume identical isolated particles sitting on an underlying lattice. Real ferrofluids are generally polydisperse systems consisting of particles with different sizes and forms. Also, real ferrofluids are much more complex and contain other internal structures in addition to isolated particles. Many experiments, computer simulations and theoretical studies of macroscopic rheological properties [22, 23] point out the existence of two other types of internal structures in ferrofluids: i) linear chain-like aggregates, and ii) bulk-drop aggregates. Any attempts for a more realistic description of ferrofluid systems by incorporating such additional realistic features will invariably lead to cumbersome physics at the expense of the simplicity invoked in this work. The focus of attention of this work is on main qualitative features of a given simple model. Thus the obtained results should be considered as qualitative, not quantitative benchmarks for the characterization of real ferrofluid systems.

III. MEAN FIELD THEORY

An exact calculation of the free energy and partition function for the lattice gas model under consideration is not possible. A common route to circumvent such difficulty when studying ordering phenomena and phase transitions is to adopt a mean field theory approach. In this context, our main interest is to search for signatures of phases that posses some type of lamellar (or stripe) order. The quantity, $\langle n(\vec{r}) \rangle$ characterizes such long range order and, as usual, $\langle \ldots \rangle$ means thermal statistical average. Since we are interested on the emergence of lamellar or striped structures parallel to the magnetic field, the simplest assumption we can make is to consider $\langle n(\vec{r}) \rangle$ a periodic function in a direction perpendicular to $\vec{B}$, namely:

$$
\langle n(\vec{r}) \rangle = \pi(x, y) = n + f(x, y) : \sum_{x, y} f(x, y) = 0, \tag{5}
$$
where \( n \) represents some uniform particle concentration and \( x, y \) are coordinates on a plane perpendicular to \( \vec{B} \). The quantity, \( \pi(x, y) \) represents the average value of particle concentration along the line of lattice sites in the \( z \)-direction, while holding \( x \) and \( y \) coordinates fixed. The periodic function, \( f(x, y) \) represents the order parameter of such system. The equilibrium state of the system is found by minimizing the free energy, \( F = E - TS \) where \( E = \langle H \rangle \) is energy, \( T \) is temperature and \( S \) is entropy. It follows from Eq.(5) that the number of occupied positions (therefore particles) along the line of lattice sites in the \( z \)-direction, while holding \( x, y \) fixed, is equal to \( N_z \pi(x, y) \) where \( N_z \) is the number of lattice sites in the \( z \) direction. The number of ways to arrange \( N_z n_{xy} \) particles on \( N_z \) available lattice sites along this line, \( W(x, y) \) is easy to calculate from well known combinatorial formulas. The number of ways to distribute all particles, \( \sum_{x,y} \pi(x, y) N_z = n N \) on \( N \) available lattice sites while satisfying the constrain in Eq.(5) is given by the product, \( W = \prod_{x,y} W(x, y) \). As suggested by standard practice in statistical physics, one applies Stirling’s formula to calculate entropy, \( S = k_B \ln W \) and obtains:

\[
S \frac{N_z}{N} = -k_B \sum_{x,y} \{ \pi(x, y) \ln \pi(x, y) + [1 - \pi(x, y)] \ln [1 - \pi(x, y)] \} .
\]  

(6)

Within the mean-field approximation one does not takes fluctuations into account and replaces \( n(\vec{r}_i) \) by its mean value \( \langle n(\vec{r}_i) \rangle \). This leads to the following expression for energy:

\[
E = J \sum_{x,y} (\pi(x, y) \pi(x + \Delta x, y) + \pi(x, y) \pi(x, y + \Delta y)] - 2 J \sum_{x,y} \pi(x, y)^2 - \mu n N B .
\]  

(7)

Putting all together, the expression for free energy, \( F = E - TS \) becomes:

\[
F \frac{N_z}{N} = \sum_{x,y} \left\{ J \left[ f(x, y) f(x + \Delta x, y) + f(x, y) f(x, y + \Delta y) \right] - 2 J f(x, y)^2 \right\} + k_B T \sum_{x,y} \left\{ \left[ n + f(x, y) \right] \ln \left[ n + f(x, y) \right] + \left[ 1 - n - f(x, y) \right] \ln \left[ 1 - n - f(x, y) \right] \right\} - \mu n N B .
\]  

(8)

The equilibrium concentration of particles along the \( z \)-direction of lattice lines as function of coordinates \( x \) and \( y \) is obtained by minimizing the free energy with respect to function \( f(x, y) \) under the constraint \( \sum_{x,y} f(x, y) = 0 \). The simplest way to describe an inhomogeneous phase that differs from a homogeneous particle concentration state is to consider a function \( f(x, y) \) that varies as a plane wave:

\[
f(x, y) = A \cos(k_x x + k_y y) ,
\]  

(9)

where \( |A| \ll n \) is some small amplitude and \( k_{x,y} \neq 2 \pi/a \). Amplitude, \( A \) represents the smallness parameter. We then proceed to expand the free energy up to fourth order in smallness parameter, \( A \). Summing over \( x \) and \( y \) we obtain:

\[
F = F_0 + A^2 \left\{ \frac{J}{2} \left[ \cos(k_x a) + \cos(k_y a) \right] + \frac{k_B T}{4 n (1 - n)} - J \right\} \cdot (1 + \alpha_2) + \frac{A^4 k_B T}{24} \left[ \frac{1}{(1 - n)^2} - \frac{1}{n^2} \right] \alpha_3 + \frac{A^4 k_B T}{96} \left[ \frac{1}{(1 - n)^3} + \frac{1}{n^3} \right] (3 + 4 \alpha_2 + \alpha_4) + O(A^4) ,
\]  

(10)

where \( F_0 = k_B T \left[ n \ln(n) + (1 - n) \ln(1 - n) \right] - \mu n B \). The set of \( \alpha \)-coefficients is: \( \alpha_i = \delta(i k_x, 2 \pi p_1/a) \cdot \delta(i k_y, 2 \pi p_2/a) \) where \( p_1, p_2 \) are arbitrary integers and \( \delta(x, y) \) is the \( \delta \)-Kronecker function. For given values of \( k_x, k_y \) we minimize free energy with respect to \( A \).
IV. RESULTS AND DISCUSSION

To begin with, we consider \( k_x = k_y = \pi/a \) and minimize the expression for the free energy, \( F/N \) in Eq.(10) with respect to amplitude, \( A \) (which represents the deviation from uniform particle concentration). We find that for temperatures which are above a critical temperature:

\[
k_B T_c = 8 J n (1 - n) ,
\]

the phase with the homogeneous spatial particle distribution (\( A = 0 \)) is stable and corresponds to the minimum of free energy. However, for temperatures below \( T_c \) the phase with modulation of the particle concentration, \( A \neq 0 \) has the lowest free energy. For this case, we calculate:

\[
A = \sqrt{3 n (1 - n) C(n)} \frac{(T_c - T)}{T} ; \quad T \leq T_c ,
\]

where \( C(n) = \frac{n (1 - n)}{(1 - 3n + 3n^2)} \) is a function of \( n \).

Fig. 1 shows the dependence of the critical temperature, \( k_B T_c/J \) as a function of \( n \) for \( 0 \leq n \leq 0.5 \). As expected, the smaller is the concentration of particles, the lower is the critical temperature at which the homogeneous-inhomogeneous phase transition happens. The predicted critical temperatures at various \( n \)-s are higher than the expected experimental values. For example, at \( n = 0.5 \) we have \( k_B T_c/J = 2 \) that is rather high considering the fact that the dipolar interaction parameter \( J/(k_B T) \) is rather low in most experimental studies.

This discrepancy is possibly due to thermal fluctuations which are underestimated in the lattice gas model. Another reason is the use of mean field theory approach, itself. From Monte Carlo simulation studies we know that theoretical approaches, when predicting an ordering transitions, are shown to generally overestimate the critical temperature [24].

In Fig. 2 we show amplitude, \( A \) as a function of \( T/T_c \) for two values, \( n = 0.1 \) and \( n = 0.5 \). The order parameter, \( A \) is continuous at the critical temperature. Its first derivative with respect to \( T \) is given by:

\[
\frac{\partial A}{\partial T} = -\sqrt{3 n (1 - n) C(n)} \frac{1}{T} \frac{1}{2 t^{3/2} \sqrt{1 - t}} ; \quad t = \frac{T}{T_c} \leq 1 .
\]

Clearly, the first derivative of \( A \) with respect to \( T \) diverges at \( T_C \) (when \( t \to 1 \)) meaning that this lattice model predicts a second order phase transition to a phase with lamellar-like ordering (where \( A \) is the continuous order parameter with diverging first derivative at \( T_c \)).

The average particle concentration in this density-modulated phase varies with position \( x, y \) as:

\[
\bar{n}(x, y) = n + A \cos \left( \frac{\pi x}{a} + \frac{\pi y}{a} \right) ,
\]

where \( A \) is given by Eq.(12). We note that the largest particle concentration, \( (n + A) \) is found on the parallel planes to \( z \)-axis placed in regular distances equal to
higher than a critical value and give rise to pseudocrystals. These experiments [bg] to the interparticle interactions of ferrofluids showed field-induced hexagonal lamellar volume fraction up to a critical value which is higher than in typical measurements [bgT bh] in concentrated cobalt ferrofluids. With an order parameter was constructed. Hence, the field strength reaches another critical value, the pattern changes from a hexagonal structure to a lamellar pattern who can broadly be associated with a lamellar phase. An earlier mean-field theoretical study [26] has also predicted isotropic, hexagonal and stripe phases for ferrofluid films. In this work, several models, including a lattice gas entropy model, were used to study thin-film ferrofluids in the presence of a magnetic field applied perpendicular to a thin sample. It was found that a ferrofluid can form spatially modulated phases at sufficiently high magnetic fields, including a stripe phase at given volume fractions. At sufficiently high field, where the fluctuations of the magnetic moment of the particles are small, an effective theory based only on the volume fraction as an order parameter was constructed.

Recent small-angle neutron scattering (SANS) measurements [27, 28] in concentrated cobalt ferrofluids (with volume fraction up to 6 % which is higher than in typical ferrofluids) showed field-induced hexagonal lamellar ordering that might be similar to the predicted lamellae. In these experiments [27], the interparticle interactions induced by an applied magnetic field dominate at fields higher than a critical value and give rise to pseudocrystalline lamellar structures in which particles are arranged in hexagonal planes, with the magnetic moments aligned parallel to the [110] direction. Molecular dynamics simulation results [20, 29] have shown hexagonal lamellar-like structures in fluids containing particles with perfectly oriented magnetic moments.

A mechanism involving competition between attractive depletion forces and repulsive interactions has been recently invoked to explain molecular dynamics simulation results in a system composed of monodisperse particles which show the stabilization of a stable lamellar phase at high volume fractions [30].

Thus, an interesting result of this work is the fact that, despite its simplicity, the lattice gas model considered here (representing a ferrofluid system in a strong magnetic field) is able to predict a second order phase transition into a phase with lamellar (stripe) spatial ordering occurring below a certain critical temperature. In this spatial-ordered phase, the largest concentration of ferrofluid particles is on planes parallel to the direction of the magnetic field. Time-resolved small angle neutron scattering experiments [17] indicate that interactions between fluctuating parallel chains aligned in a magnetic field lead to lateral attraction thus lamellar ordering. While unrelated, this type of ordering is reminiscent of stripe-like or liquid crystalline ordering recently seen in certain quantum Hall regime electronic systems [31].

It is possible that other heterogeneous phases are the thermodynamically stable states. Typically, chain-like structures are predicted theoretically, observed in simulations, and seen in experiments. Some authors have predicted phase transitions at sufficiently low temperatures [32] from chain-like structures to heterogeneous structures of other nature. In this scenario, the phase transitions begins with the formation of linear chains and collapse of longest of them into dense compact globules. Evolution of the globules leads to equilibrium separation of ferrofluid into two phases, with high and low concentration of particles [32]. Thus, it is possible that the lattice gas model considered in this work, at least at a qualitative level, might be related to such a scenario, too.

In conclusion, we presented a simple lattice gas model for a ferrofluid of magnetic particles with their magnetic moments fully aligned in the direction of a strong external magnetic field. We assumed only nearest-neighbor dipole-dipole interaction between particles placed at sites of a simple cubic lattice. Within a mean field approximation we found a second order phase transition from a phase with homogeneous spatial particle distribution to a phase with periodic positional ordering. In this approximation, the spatial period of the arising structure does not depend on the particle concentration. In order to simplify the problem, we studied only the stability of lamellae-like structures. The theoretical study of other than lamillae heterogeneous structures (for instance chain-like structures, etc) has not been considered and merits a separate analysis. Therefore, at present, we can only conclude that: (i) spatial structures stabilize

<table>
<thead>
<tr>
<th>$k_x$</th>
<th>$k_y$</th>
<th>$k_B T_c$</th>
<th>$\lambda$</th>
<th>$F/N$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\pi/a$</td>
<td>$\pi/a$</td>
<td>$8 J n (1 - n)$</td>
<td>$\sqrt{2} a$</td>
<td>$F_0 - \frac{2 k_B (T - T_c)^2}{8 a^3} C(n)$</td>
</tr>
<tr>
<td>$\pi/(4 a)$</td>
<td>$\pi/(4 a)$</td>
<td>$4 J n (1 - n)$</td>
<td>$2\sqrt{2} a$</td>
<td>$F_0 - \frac{4 k_B (T - T_c)^2}{8 a^3} C(n)$</td>
</tr>
<tr>
<td>$\pi/(6 a)$</td>
<td>$\pi/(6 a)$</td>
<td>$2 J n (1 - n)$</td>
<td>$3\sqrt{2} a$</td>
<td>$F_0 - \frac{6 k_B (T - T_c)^2}{8 a^3} C(n)$</td>
</tr>
</tbody>
</table>

TABLE I: Some lamellar-like structures that minimize the free energy: $\lambda = \sqrt{2} a$ - stable phase; $\lambda = 2\sqrt{2} a$ - metastable phase; and $\lambda = 3\sqrt{2} a$ - metastable phase. The lattice parameter of the underlying simple cubic lattice is $a$ and $C(n)$ is a function of particle concentration, $n$. 

$\sqrt{2} a$. This spatial structure may be crudely identified with lamellar-like ordered structures found in some ferrofluids under high magnetic fields. Other lamellar phases arise if one repeats the same free energy minimization procedure (with respect to $A$), but with different sets of fixed $k_x$ and $k_y$ parameters. Some results obtained when $k_x = k_y = \pi/(4 a)$ and $k_x = k_y = \pi/(6 a)$ are shown in Table I (third and fourth row). The third column in Table I represents the values of the critical temperature, $T_c$ below which lamellar phases possibly stabilize. The parameter, $\lambda$ in the fourth column of Table I is the distance between two adjacent planes each with maximum particle concentration (when $n_{xy} = n + A$).

It is of interest to compare these results with experiments or computer simulations. For instance, experiments in Ref. [25], despite being done in thin films, indicate the appearance of labyrinthine patterns for high perpendicular magnetic fields. In a weak perpendicular magnetic field, columns parallel to the direction of the applied field are formed. By increasing the magnetic field, a hexagonal crystal structure appears, but when the field strength reaches another critical value, the pattern changes from a hexagonal structure to a lamellar pattern who can broadly be associated with a lamellar phase. An earlier mean-field theoretical study [26] has also predicted isotropic, hexagonal and stripe phases for ferrofluid films. In this work, several models, including a lattice gas entropy model, were used to study thin-film ferrofluids in the presence of a magnetic field applied perpendicular to a thin sample. It was found that a ferrofluid can form spatially modulated phases at sufficiently high magnetic fields, including a stripe phase at given volume fractions. At sufficiently high field, where the fluctuations of the magnetic moment of the particles are small, an effective theory based only on the volume fraction as an order parameter was constructed.

Recent small-angle neutron scattering (SANS) measurements [27, 28] in concentrated cobalt ferrofluids (with volume fraction up to 6 % which is higher than in typical ferrofluids) showed field-induced hexagonal lamellar ordering that might be similar to the predicted lamellae. In these experiments [27], the interparticle interactions induced by an applied magnetic field dominate at fields higher than a critical value and give rise to pseudocrystalline lamellar structures in which particles are arranged in hexagonal planes, with the magnetic moments aligned parallel to the [110] direction. Molecular dynamics simulation results [20, 29] have shown hexagonal lamellar-like structures in fluids containing particles with perfectly oriented magnetic moments.

A mechanism involving competition between attractive depletion forces and repulsive interactions has been recently invoked to explain molecular dynamics simulation results in a system composed of monodisperse particles which show the stabilization of a stable lamellar phase at high volume fractions [30].

Thus, an interesting result of this work is the fact that, despite its simplicity, the lattice gas model considered here (representing a ferrofluid system in a strong magnetic field) is able to predict a second order phase transition into a phase with lamellar (stripe) spatial ordering occurring below a certain critical temperature. In this spatial-ordered phase, the largest concentration of ferrofluid particles is on planes parallel to the direction of the magnetic field. Time-resolved small angle neutron scattering experiments [17] indicate that interactions between fluctuating parallel chains aligned in a magnetic field lead to lateral attraction thus lamellar ordering. While unrelated, this type of ordering is reminiscent of stripe-like or liquid crystalline ordering recently seen in certain quantum Hall regime electronic systems [31].

It is possible that other heterogeneous phases are the thermodynamically stable states. Typically, chain-like structures are predicted theoretically, observed in simulations, and seen in experiments. Some authors have predicted phase transitions at sufficiently low temperatures [32] from chain-like structures to heterogeneous structures of other nature. In this scenario, the phase transitions begins with the formation of linear chains and collapse of longest of them into dense compact globules. Evolution of the globules leads to equilibrium separation of ferrofluid into two phases, with high and low concentration of particles [32]. Thus, it is possible that the lattice gas model considered in this work, at least at a qualitative level, might be related to such a scenario, too.

In conclusion, we presented a simple lattice gas model for a ferrofluid of magnetic particles with their magnetic moments fully aligned in the direction of a strong external magnetic field. We assumed only nearest-neighbor dipole-dipole interaction between particles placed at sites of a simple cubic lattice. Within a mean field approximation we found a second order phase transition from a phase with homogeneous spatial particle distribution to a phase with periodic positional ordering. In this approximation, the spatial period of the arising structure does not depend on the particle concentration. In order to simplify the problem, we studied only the stability of lamellae-like structures. The theoretical study of other than lamillae heterogeneous structures (for instance chain-like structures, etc) has not been considered and merits a separate analysis. Therefore, at present, we can only conclude that: (i) spatial structures stabilize...
within the lattice gas model at low temperatures; and (ii) lamellae structures are possible candidates for these structures.

In future work, we plan to further analyze the phase diagram in order to discriminate among many other competitive states, like small chains of particles and/or arrays of long chains of particles all oriented along the direction of the magnetic field.

Acknowledgments

This research was supported in part by the National Science Foundation under Grant No. PHY05-51164.