Honors Projects   Physics

5-1993

A Lattice Gas Approach to the Structure and Dynamics of Electrorheological Fluids

Jie Chen '93
Illinois Wesleyan University

A Lattice Gas Approach to the Structure and Dynamics of Electrorheological Fluids

A Dissertation Submitted

by

Jie Chen

to
The Department of Physics

In partial Fulfillment of the requirements for

Research Honors in Physics

at

Illinois Wesleyan University
Bloomington, IL 61702-2900

May 1993

Advisor: Dr. Narendra K. Jaggi

A Lattice Gas Approach to the Structure and Dynamics of Electrorheological Fluids

Jie Chen *

Department of Physics
Illinois Wesleyan University
Bloomington, IL 61702

Synopsis

Electrorheological fluids consist of a colloidal suspension of dielectric particles in a continuous fluid of smaller dielectric constant. Molecular dynamics simulations of these fluids in an applied electric field have recently been shown to produce percolated, columnar structures. No systematic attempt has been made so far to simultaneously include the effects of temperature and the viscous drag due to the continuous fluid. We propose a dipolar lattice gas model for electrorheological fluids and study the resulting structures and dynamics. We attempt to incorporate the effect of the viscosity of the continuous medium by a dynamic ansatz that determines the range over which individual particles can jump in a single simulation event. The temperature is simulated by assigning a probability of jumping to higher energy states in accordance with the Boltzman distribution. We study the equilibrium phases of the system as a function of temperature and find interesting new results. Our new results from finite temperature simulations suggest that there is a gradual phase transformation from a liquid like phase at low electric field or equivalently at high temperature to a solid like phase at high electric field or at low temperature. The simplicity obtained by going to a lattice version will allow us to extend out these simulations even in three dimensions, where little is known about these systems.

1   Introduction

Electrorheological (ER) fluids are a class of novel materials that have received considerable attention in the scientific community recently [1, 3, 5, 6]. They consist of small highly polarizable solid particles suspended in low dielectric constant fluids. One interesting characteristic of this fluid is that when an electric field is applied across it, its viscosity increases almost immediately by a very large amount. It behaves more like a weak solid than a liquid. When the electric field is removed, it reverts back to its liquid state almost instantly. Because it is easy and inexpensive to generate electric fields and the response time of this fluid to the applied electric fields is of the order of 10^{-3} second, this interesting property has many potential applications, such as the next generation of low cost active automotive suspension, brake and transmission systems.

2   The Problem

The fundamental physics of this phenomenon needs to be understood before engineers can take full advantage of these interesting materials. In particular,

1. What does the structure of ER fluids look like in an applied electric field?

2. Why can it change from a liquid to a weak solid? What types of forces are involved?

3. How does the system evolve in time?

4. What are the effects of temperature on the equilibrium states of the ER fluids? Are there any phase transitions, as the electric field is increased or the temperature is decreased?

3   A Summary of Past Work

Since the particles suspended in the liquid continuous medium are relatively large (1-5 μm), the structure of the ER fluids can be studied directly
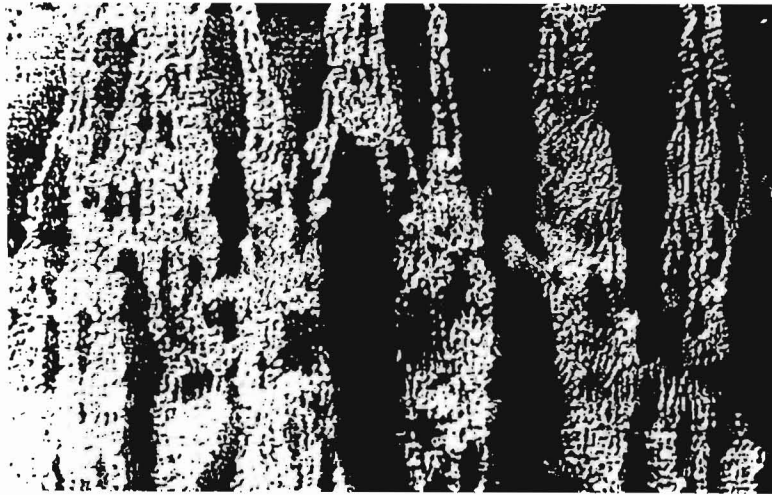
1

Figure 1. Structure of ER Fluids
By Whittle, 1989 [1]

by optical microscopy. Figure 1 shows a recent result by Whittle [1]. We note that the particles cluster and form chain like structures along the direction of electric field. These chain like structures are not simple straight lines, but intertwined and twisted. Also, the width of the columns changes significantly as one moves along the field direction. As a result of this columnar structure, the viscosity of the fluids increases as the chains become longer and thicker.

During the last five years, a consensus seems to have emerged that there are basically three types of forces that dominate the physics during the transformation [2]. They are:

- The induced dipolar interactions among these small particles;

- The viscous drag forces on the particles as they move through the continuous medium;

- The thermal fluctuation of the system at finite temperatures.

Although we are quite clear on the types of forces involved, a complete understanding is not yet available. This is a many-body problem and the

forces involved are quite complex: they are long range and anisotropic. It has not been possible to solve the problem analytically. As a matter of fact, most of the progress in understanding these complex fluids has been made by computer simulations [3, 4, 5]. The results of these computer simulations are in pleasant agreement with the structures obtained experimentally. But there are still some interesting questions that have not been explored, such as:

1. No systematic attempt has been made to include both the effects of finite temperature and viscous drag due to the continuous medium;

2. Previous simulations use numerical methods to solve a system of coupled differential equations derived from the classical Newtonian mechanics. Because of their computational complexity, they can only simulate a small number of particles. But since the dipolar interaction is a long range interaction, the boundary effects may shadow the results in these limited simulations. There is a need to study larger systems to consolidate our understanding of this material.

The organization of the rest of the paper is as follows. **Section 4** presents a new discrete approach: *the Dipolar Lattice Gas Approach* to study the *structure, dynamics and thermodynamics* of the ER fluids. Both the zero temperature and finite temperature situations are studied. Because the particles are restricted on a discrete lattice, it has significant computational savings over the continuous models. As we will see the results in **Section 5**, this approach not only generates structures in reasonable agreement with the previous experimental and simulation results, it also enables us to address the above unanswered questions and even go to three-dimensions, where little is known about these systems.

4   Dipolar Lattice Gas Approach

Previous computer simulations use continuous models where the particles can move freely on a continuum according to Newtonian mechanics. We restrict the positions of the particles on a lattice grid. The particles can only occupy the intersections of the lattice and no two particles can occupy the same position. In this paper, we focus on the two dimensional square lattice.

3

Figure 2.   Lattice Grid
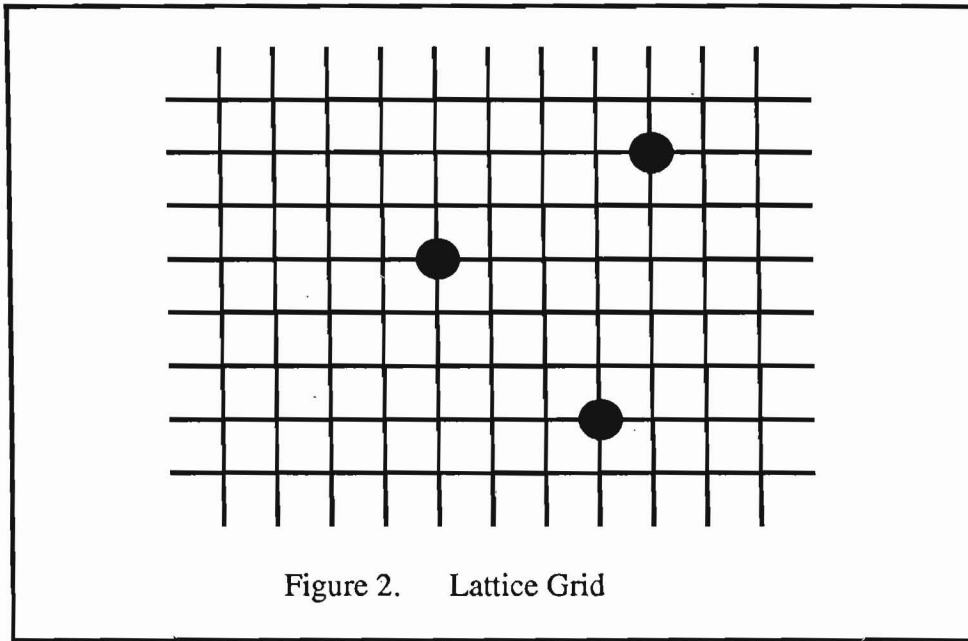
There are fluctuations in local density, but the overall density of the system is held constant throughout the transformation. Since there is a repulsive component in the dipolar interaction, we introduce the following boundary conditions to assure that the overall density remains constant. Whenever a particle moves out of the left boundary, it is introduced back to the corresponding position on the right side, as if the lattice grid were circular in the horizontal direction. The same prescription is applied to particles moving out of the right boundary. Since the up and down boundaries represent the two metallic electrodes, the particles are not allowed to jump out of the up and down boundaries. Mirror image boundary conditions are applied, as required by the theories in electromagnetism.

4.1   Structure and Dynamics at Zero Temperature

The system begins with a certain density of randomly placed particles on the lattice. As an electric field is applied across them, they become induced dipoles in the direction of the external electric field. The electrostatic
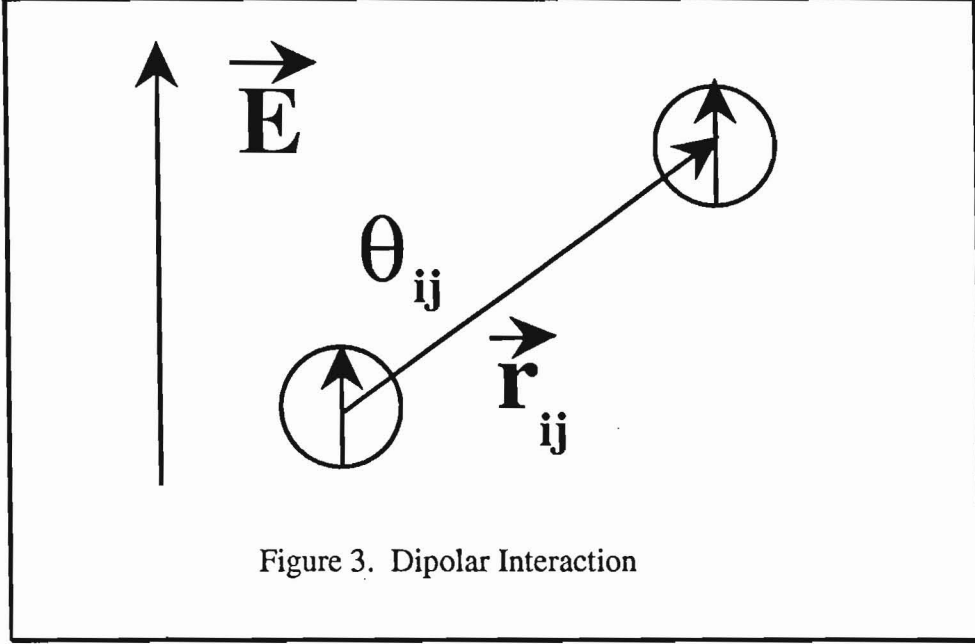
4

Figure 3. Dipolar Interaction

force $\vec{F_i}$ on the ith dipole (See Figure 3) is characterized by

$$
\begin{aligned}
\vec{F_i} &= -\nabla_i U(\{\vec{r_j}\}) \\
&= 3p^2 \sum_j \frac{1}{r_{ij}^4}[(3\cos^2\theta_{ij} - 1)\hat{e}_r + (\sin 2\theta_{ij})\hat{e}_\theta],
\end{aligned}
\tag{1}
$$

where p is the induced dipolar moment in the direction of the applied electric field; r_{ij} is the distance between particles i and j; θ_{ij} is the angle between vector $\vec{r}_{ij}$ and the direction of the electric field; and the energy of the system $U(\{\vec{r_j}\})$ is given by the following equation:

$$
U(\{\vec{r_j}\}) = -p^2 \sum_{\{ij\}} \frac{(3\cos^2\theta_{ij} - 1)}{r_{ij}^3}
\tag{2}
$$

with the sum runs over all dipoles and their sequence of images in the two metallic electrodes.

At zero temperature, because of the viscous drag force, the system will evolve in the direction of decreasing energy. Let us first define a parameter
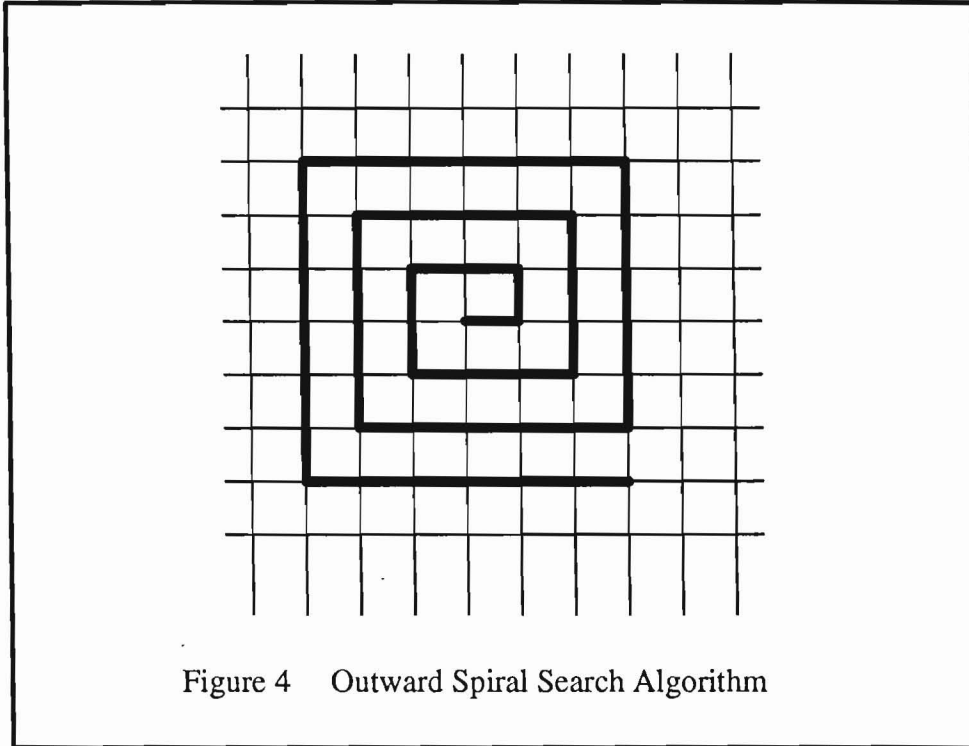
5

Figure 4   Outward Spiral Search Algorithm

called "neighborhood". The "neighborhood" of a particle is defined as a square area, with the particle at its center, within which the particle can attempt to jump to in a single move. We select a particle at random and use the following procedure to determine where, if at all, the particle will move to during this interation.

A target empty lattice site is selected within the "neighborhood". If this tentative move can lower the total energy, we accept it and repeat the procedure on another randomly selected particle. If it cannot lower the total energy, we go on to try another empty site in the "neighborhood", until all the empty sites within the "neighborhood" have been exhausted. The whole procedure is iterated again until we stop it.

How shall we choose an empty site within the neighborhood to make a tentative move? Our initial attempt was to check all possible sites within the specified neighborhood and find the one that can lower the total energy the most, and move the particle to that site. This method does give us the

steepest decrease in energy. However, it needs to check every possible site in the neighborhood; it is computationally too expensive as we go to larger lattices.

Another way to select a target site is to move as soon as it finds a site that can lower the energy. This scheme raised a question: what should be the search order? We want to be sure that the final structure of the system is the result of the underlying physics, but not due to the way an algorithm is implemented. If we search the new position in the neighborhood starting from the north-west corner sequentially down to the south-east corner, there is a tendency for particles to cluster in the north-west corner. After rethinking the physics, we adopted a dynamic ansatz which we call *Outward Spiral Search Algorithm*. It is natural to think that we shall first check the near neighborhood of the particle. If sites in the near neighborhood cannot lower the energy, then we search for outer neighborhood sites.

Since the dipolar interaction decreases as $\frac{1}{r^3}$, the farther away a particle, the less its influence on others. We introduced a "range" parameter as the cut off during the summation of the interactions. It is also specified as a square area. If a particle sits outside the range-square centered at the current particle, their mutual interaction is ignored.

We hope that the viscosity of the continuous medium is simulated by the size of the neighborhood. Larger neighborhood size will correspond to less viscous liquid medium.

One implementation detail is worth mentioning here. Our formal criterion for moving a particle to a new site is related to the total energy, which is a double summation over all the particles. In pratice, however, we only need to compute a partial energy U_i, which is a single summation, as follows

$$U_i = -p^2 \sum_{\{j \neq i\}} \frac{(3\cos^2 \theta_{ij} - 1)}{r_{ij}^3}, \tag{3}$$

here U_i is the sum of interaction energies of the particle that we are attempting a move with the other particles in the system. Since we move one particle at a time, the relative positions of the other particles are unchanged. The energy associated with those particles does not change either. In order to determine whether the new move can lower the total energy, we need only to know if the move can lower the partial energy associated with the particle itself. This reduction of a double summation to a single summation results in significant computational savings.

Figure 5 .   Simulation of Thermal Effects
of Using Finite Temperature Boltzman Distribution
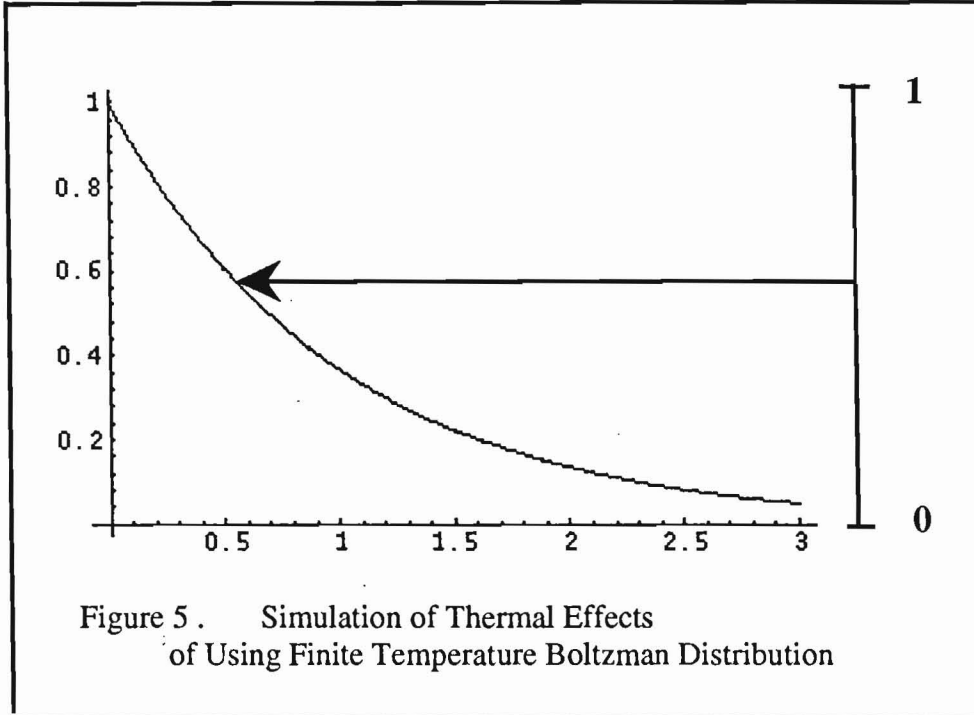
4.2   Equilibrium Thermodynamic States at Finite Temperature

There is a standard way to incorporate the temperature effects on the equilibrium states of the system. We use the symbol ΔU to represent the energy difference before and after the tentative move, k as the Boltzman constant and T as the temperature. Statistical mechanics tells us that when $\Delta U > 0$, the system has a finite probability of occupying these higher energy states according to the Boltzman distribution, given by

$$P(\Delta U) = e^{-\frac{\Delta U}{kT}}. \tag{4}$$

For a fixed positive ΔU, the higher the temperature, the larger the probability that it can jump to the higher energy state.

We guarantee this distribution in the simulation by the following procedure. After a tentative move, ΔU is calculated. If $\Delta U < 0$, the tentative move is accepted. If $\Delta U > 0$, the probability factor $P(\Delta U)$ at this ΔU

8

is calculated. A random number is drawn from a uniform random number generator. If the random number is smaller that the calculated probability factor $P(\Delta U)$, the tentative move is accepted; otherwise, the tentative move is rejected. After many trials, collectively, the frequency of accepting moves to higher energy states is in accordance with the Boltzman distribution.

5 Results and Conclusions

The electric field is turned on at $t = 0$. At each specified time interval, the total energy of the system is calculated, and the energy and the structure of the system are logged into files. By running the simulation for a long time, we are able to study the structure, dynamics and thermodynamics of the systems.

5.1 Structure and Dynamics at Zero Temperature

At zero temperature, the total energy of the system decreases monotonically. Figure 6 is a plot of total energy vs. time of a 100 x 100 lattice. Figure 7 gives us the structure of the system after a considerable long time (t=11 units). As expected, we see long chains formed along the direction of electric field between the two electrodes. Also, we can see intertwined connections between chains of various thickness. We can see the transformation of the structure of the system by looking at several snapshots at time 0, 2 and 11 units (Figure 8). The systems begins with a random configuration. After an electric field is applied across it, particles cluster to form short chains. At time 2, we can see that the chain like structures are becoming longer in the direction of the electric field. As the system evolves, the columnar structure becomes longer and thicker. At time 11, the columnar structures completely span the two electrodes and all intertwined together.

We are pleased to see that our new approach generates results with resonable agreement with the previous experimental and computational works. This gives us the confidence to go beyond the structure and dynamics at $T = 0$, and study the thermodynamic properties of the ER fluids at finite temperatures, which has not been explored earlier.
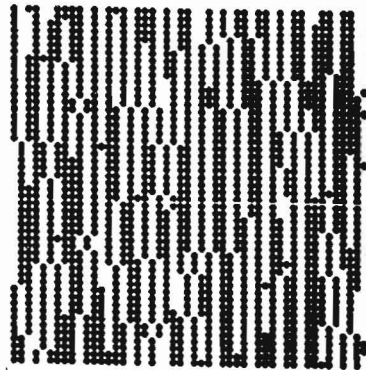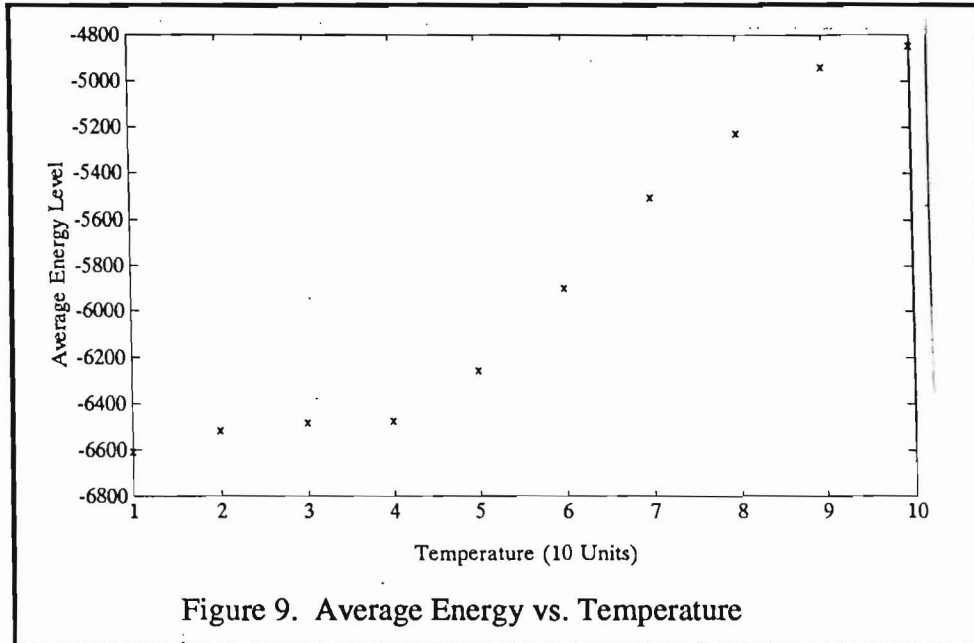
Figure 6. Total Energy vs. Time
(At zero temperature)

Figure 7. Structue after a Long Time (t=11)

10
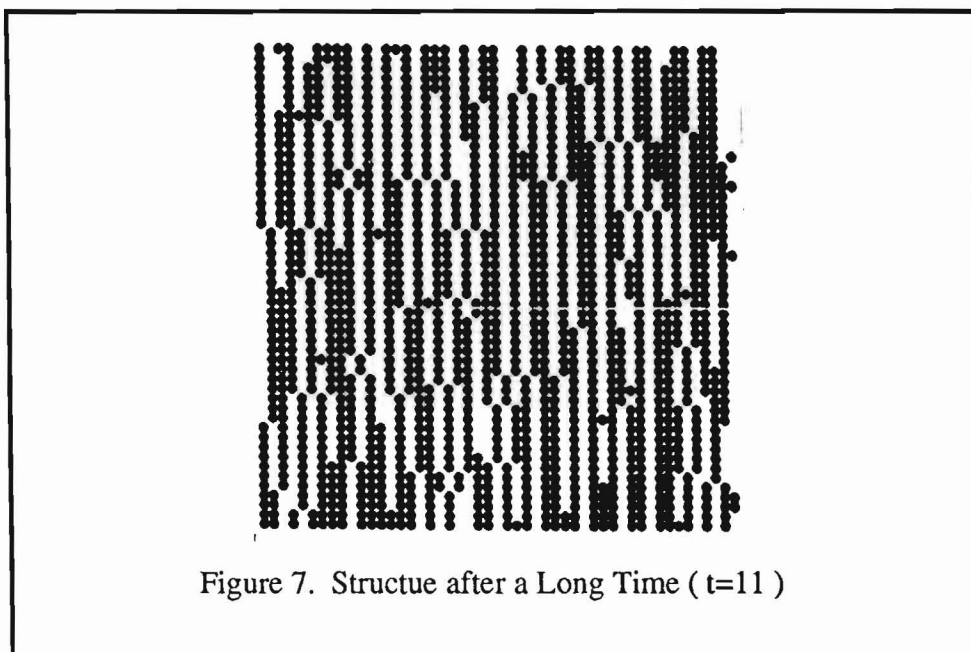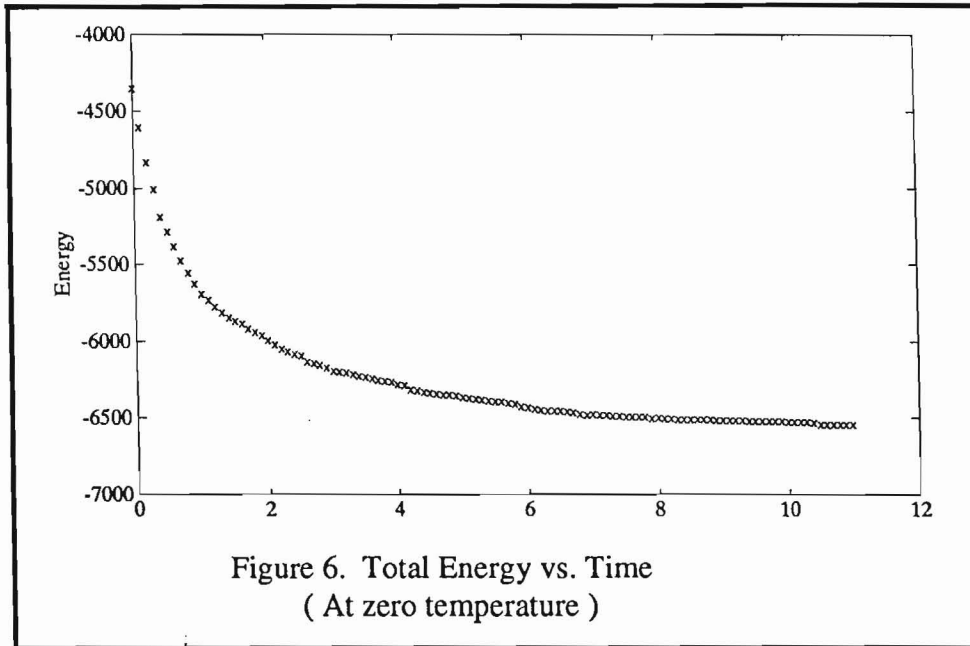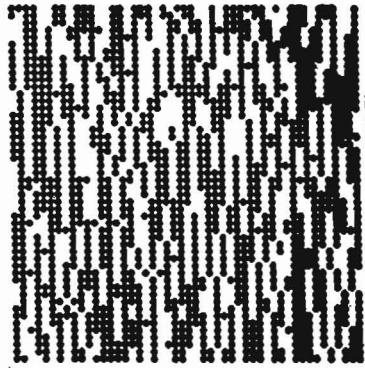
Figure 8 Three Structures at t=0, 2, 11
(At zero temperature)

t = 0

t = 2

t = 11

Figure 9. Average Energy vs. Temperature

5.2   Temperature Effects on the Equilibrium States

At finite temperature, the particles have certain probabilities to jump to higher energy states. Since the system is continuously changing, there is no single final structure for it. But after a sufficiently long time, the system will reach its thermodynamic equilibrium state. At equilibrium state, the structure and the energy of the system still fluctuate around its average structure and energy. But at a fixed temperature, the average total energy of the system over a long time is a constant. Figure 9 is a plot of the average total energy vs. temperature. There is a gradual transition from low average energy at low temperature to high average energy at high temperature. Figure 10 presents some typical structures of the system at these different temperatures (T=2,4,6,8,10). At low temperature, the equilibrium state of the system is well ordered, which is like a solid. While at high temperature, the columnar structures are broken up . The structure of the system is very disordered, or more liquid like. Also we can see a gradual transition of the
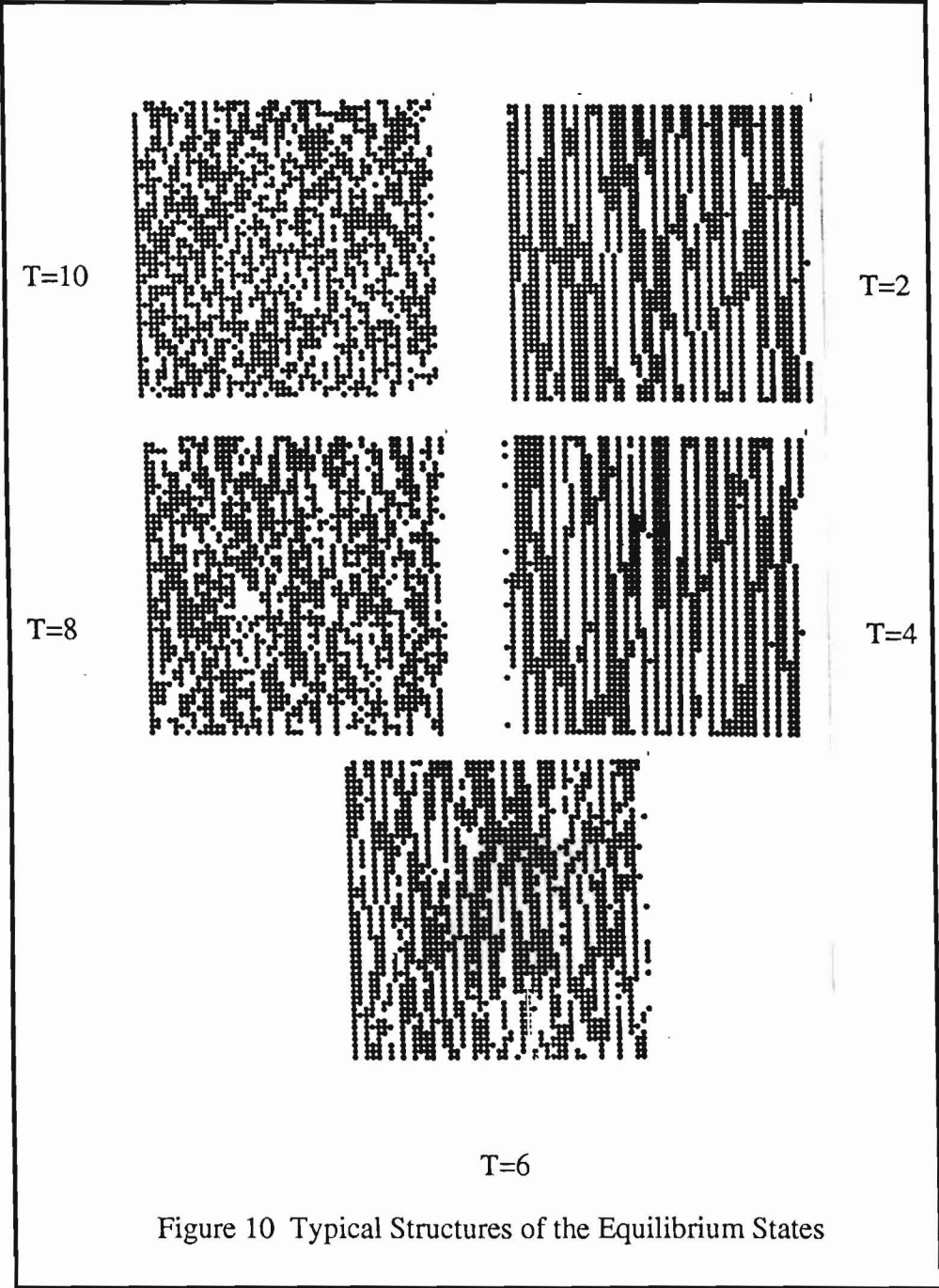
T=10

T=2

T=8

T=4

T=6

Figure 10 Typical Structures of the Equilibrium States

structure from well ordered to disordered as temperature increases, which correlates with the gradual transition of the average energy.

Since the temperature effects are simulated by assigning probabilities of jumping to higher energy states according to the Boltzman distribution, what matters is the ratio of energy difference to temperature. So high electric field strength is equivalent to low temperature, and vice versa.

To summarize, we see a gradual transition of the system from a solid like phase to a liquid like as the temperature increases or equivalently, as the strength of the electric field decreases. There is no sharp transition like the one that occurs in a true "phase transition". This is in agreement with almost all the experimental results [3, 6]. The thermodynamic properties of the system can be further studied by analyzing the fluctuations of the energy in its equilibrium states.

6   Acknowledgements *

References

[1] M. Whittle, *Physics World* 2 (1989) p39

[2] A.P.Gast and C.F. Zukoski, *Advance in Colloid and Interface Science* 80 (1989) p153

[3] D.J. Klingenberg, F. van Swol, and C.F. Zukoski, *Journal of Chemical Physics* 91 (1989) p7888

[4] R.T. Bonnecaze and J.F. Brady, *Electrorheological Fluids: Proc. Second Int. Conf. on ER Fluids* (1990) p27

[5] N.K. Jaggi, *Journal of Statistical Physics* Vol 64(1991) p1093

[6] J.A.Payne and N.K. Jaggi, *Bulletin of the American Physical Society* Vol 38(1993) p396