

## ELECTRIC-FIELD-INDUCED LAMELLAR STRUCTURES IN MAGNETIC FLUIDS— A 2D DIFFUSION MODEL

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A lamellar pattern can form in a thin magnetic fluid layer when the applied electric field is above a critical value. A 2D simulation is performed to study the field-induced phase separation and the pattern by using the mass continuity equation. The simulation produces the similar structure in field but does not match the experimental growth law.

Most studies of field-induced structures in magnetic fluids have investigated the role of magnetic fields. Much less is known about the effects of electric fields applied to magnetic fluids[1]. Recent experiments have shown that a DC electric field can induce phase separation and lamellar structures in magnetic fluids[2]. In order to understand the physical mechanism for this behavior, we have carried out computer simulations on a lattice gas model, which is based on a two dimensional diffusion equation.

We begin our study from the experimental observations that the lamellar structure is observed in very thin cells with an electric field imposed perpendicular to the ferrofluid layer, and is associated with the migration of magnetic particles within the fluid[1]. In the presence of a uniform electric field  $E$ , at temperature  $T$ , the free energy density of the system can be written as

$$F = F_{bulk} + F_{inhomo}, \text{ with } F_{bulk} = U_E - TS \text{ and } F_{inhomo} = k \nabla^2 C. \tag{1}$$

Here  $U_E$  is the electrostatic energy contribution due to the electric field, and is given by:

$$U_E = - 1/2 \ \varepsilon(C) E^2, \tag{2}$$

where the dielectric constant  $\varepsilon(C)$  depends on the local particle concentration,  $C(\mathbf{r})$  in a nonlinear fashion. (The negative sign in  $U_E$  is a consequence of choosing the electric field as a thermodynamic variable.) For our choice of  $\varepsilon(C)$ , we use the expression

$$\varepsilon(C) = \exp [ C \ln \varepsilon_p + (1 - C) \ln \varepsilon_f ] \tag{3}$$

which has been used previously to study phase separation process under the influence of an electric field[2].

For the entropy,  $S$ , we use the entropy of mixing from a lattice-gas model for a binary system.

$$S = (k_B / \nu)T [ C \ln C + (1 - C) \ln (1 - C) ] \quad (4)$$

Here  $\nu$  is the average correlation volume. (For a non-interacting system,  $\nu$  is the volume of a single particle). The inhomogeneous term  $F_{inhomo}$  represents the energy of interfaces which results from phase separation.

The time-dependent Ginzburg-Landau equation for the concentration,  $C(\mathbf{r}, t)$ , can be written as

$$dC/dt = M \nabla^2 \mu, \quad (5)$$

where  $M$  is the mobility and the local chemical potential  $\mu(\mathbf{r}) = \delta F / \delta c(\mathbf{r})$  is obtained from the free energy functional above, which results in the following equation:

$$dC/dt = \nabla^2 (-R \exp(\ln(\epsilon_p/\epsilon_f) C) E^2 + \ln(C/(1-C)) - \nabla^2 C) \quad (6)$$

Both length and time are scaled by  $\lambda$  and  $\tau$  which are defined as:

$$\tau = \nu^2 K / M (k_B T)^2 \quad (7)$$

$$\lambda^2 = K \nu / k_B T, \quad (8)$$

and the control parameter  $R$  is given by:

$$R = (\epsilon_f \ln(\epsilon_p/\epsilon_f) E^2 \nu / 2 k_B T) \quad (9)$$

Temperature is held constant at  $T=298$  K. The two main variables used are voltage and time; voltage is used in place of electric field, assuming a separation between plates of  $d \approx 10 \mu\text{m}$ . For the dielectric constants we use  $\epsilon_p = 20$ , the value for magnetite particles; and  $\epsilon_f = 2$ , the approximate value for kerosene at room temperature. For the average correlation volume, we choose it with a radius of 10 times the average particle radius as found from other calculations [3]. The average concentration is set to 7% to match experiment. The surface energy constant,  $K$ , is set to one as it is included with the scaling factors of the length and time. As is apparent from the control parameter, the value of  $K$  does not affect the critical values for phase separation. However, for direct comparison with experiment, the value of  $K$  is needed to convert time and distance from scaled to real units.

The initial configuration at  $t=0$  is drawn from a random fluctuation of 1% around the mean concentration. We observe phase separation beyond a critical value of the electric field,  $E_c$ , consistent with experimental result[4, 5]. Below the critical field, the system always returns to the same original local concentrations, even if there is a momentary surge away from the average. At equal to or above the critical value, the deviation of local concentration from the average grows with time.

Figure 1 and figure 2 show the time development of the lamellar pattern. The dark/light regions represent concentrations greater/less than the average value. The

amplitude is defined as the difference between the averages of the high and the low regions. Below  $E_c$ , no phase separation occurs and the amplitude difference between the two regions is not visible. For an average concentration of 7%, the critical voltage in the simulation is 10 volts at the room temperature. At this value, lamellar structures form at time  $t \sim \tau$ , indicating that the labyrinthine pattern observed in experiment can be explained by a diffusion process.

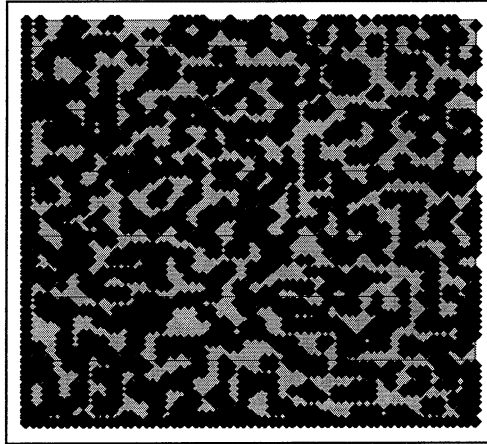


Fig. 1 Structure formation at  $t=0.25\tau$ ,  $c=7\%$ ,  $V=10$  V,  $T=298$  K

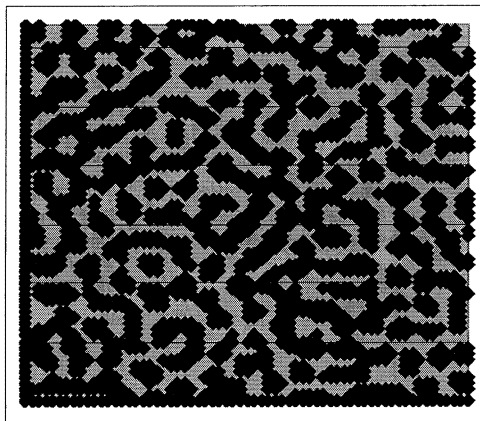


Fig. 2 Structure formation at  $t=1.5\tau$ ,  $c=7\%$ ,  $V=10$  V,  $T=298$  K

The width of the stripes increases at short time until  $t \sim \tau$ , then decreases slightly. Experimentally, the stripe width grows with time, following a power law with an exponent of  $1/3$  [6]. This may indicate that electric field is not the only driving force for the phase separation.

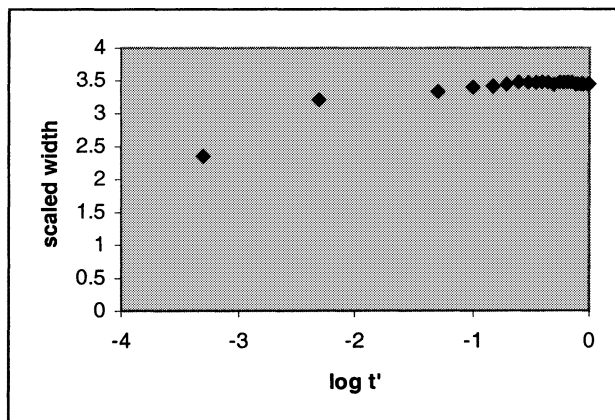


Fig. 3 Stripe width vs. time;  $c=7\%$ ,  $V=10$  V,  $T=298$  K

Future work with the simulation will include studies of varying voltage and temperature, and a direct comparison of the simulation with experiment. This work is supported by US NSF DMR under grant #0072901.

### References

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