

Classification

Physics Abstracts

75.30 — 64.75 — 82.70D — 46.60

## Structure induced in suspensions by a magnetic field

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*(Received 3 February 1993, revised 19 October 1993, accepted 9 November 1993)*

**Abstract.** — Magnetorheological as well as electrorheological fluids exhibit a phase separation when a field is applied to the suspension. We observe the formation of aggregates elongated in the direction of the field and whose length  $L$  is fixed by the thickness of the cell. The average radius  $b$  of these aggregates has been measured for  $L$  between 100  $\mu\text{m}$  and 700  $\mu\text{m}$  for two volume fractions  $\Phi = 5\%$  and  $\Phi = 10\%$ . In this range we find for a magnetorheological fluid composed of micronic superparamagnetic particles an experimental law  $b \propto a(L/a)^n$  with an exponent  $n \approx 0.67 \pm 0.04$ . We can recover this experimental law and predict the characteristic size of the aggregates from the knowledge of the magnetic permeability of the particles with a model based on the hypothesis of close packed aggregates. The agreement with the experiments is good especially at the lower volume fraction. Actually on a larger range of thicknesses our model predicts a continuous increase of this exponent towards a final value of one for large values of  $L/a$ .

### 1. Introduction.

Magnetorheological suspensions are composed of magnetic particles whose size, in the micronic range, is at least two orders of magnitude larger than in the better known magnetic suspensions, called ferrofluids. The induced magnetostatic force between two particles being proportional to the square of the radii of the particles, it is much larger for M. R. fluids than for ferrofluids. As a counterpart, sedimentation is no longer prevented by Brownian motion and the micronic range represents an optimum size. Under the application of a magnetic field [of a few Oersted] we observe a transition from a homogeneous liquid suspension to a solid one [1]. This transition from a liquid to an apparent solid state is due to the formation of elongated aggregates of particles which are aligned parallel to the magnetic field and join the opposite sides of the cell containing the suspension. These links are responsible for the apparent solid behavior since, for instance, if the field is perpendicular to the two disks of a rheometer, we need to increase the imposed torque above a critical value in order to break these aggregates

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(\*) CNRS - URA 190.

and obtain a continuous rotation of the upper disk. The onset of a yield stress strongly modifies the rheological behavior and opens the possibility to realize hydraulic devices where the viscosity is electronically controlled. This effect, due to the magnetization of the solid particles by a magnetic field can, as well, be obtained with dielectric particles polarized by the application of an electric field. These electrorheological fluids are the object of a rising interest both for their potential applications and for the interesting underlying physics [2-4]. Clearly we need to obtain a good knowledge of the structure of these suspensions — the shape of the aggregates, the distance between them, the internal arrangement of the particles — if we want to be able to predict the change of their rheological properties in the presence of a field.

For monodisperse dielectric particles, Tao *et al.* [5, 6] predict and observe an internal structure for these aggregates composed of chains of spheres (of radius  $a$ ) with a given chain surrounded by four other ones at a distance  $a\sqrt{3}$  with a shift of a radius in the direction of the applied field and four next-nearest neighbors at a distance  $a\sqrt{6}$ , but without shift. Such a configuration called B.C.T. (body centered tetragonal) gives the lowest internal energy. In a recent paper, Halsey and Toor [7] model the aggregate as a spheroidal droplet and calculate the internal energy on the basis of electrostatics of continuous media. They furthermore introduce a surface energy  $U_\sigma$  proportional to the square of the applied field  $E$ :

$$U_\sigma \approx 0.1 \beta^2 E^2 a S_a \quad \text{with} \quad \beta = \frac{\varepsilon - 1}{\varepsilon + 2} \quad (1)$$

$S_a$  is the surface of the aggregate,  $a$  the radius of a particle and  $\varepsilon$  the ratio of the permittivity of the solid particle to that of the suspending fluid. In this approximation  $U_\sigma$  is independent of the shape of the aggregate. Minimizing the total energy they obtain, in the limit where the ratio of the equatorial semi-axis to the total length  $b/L$  tends to zero, a dependence represented by  $b \propto a(L/a)^{2/3}$ . In this model the energy of interaction between different aggregates is not taken into account. Actually when the aggregates fill the gap between the electrodes, the existence of image dipoles which impose the potential on the electrodes is equivalent to the introduction of aggregates of infinite length. Then, at this last stage of aggregation, the depolarization field and consequently the repulsive interaction between the aggregates disappears.

We expect the surface energy to be given by the same expression for magnetic and for dielectric particles: we just have to replace  $\varepsilon$  by  $\mu$ , the magnetic permeability, and  $E$  by  $H$ , the average magnetic field inside the sample. This will be true as long as the permeability does not vary too much with the magnetic field. In a previous work [1] we have reported experimental results about the average size of these aggregates *versus* the initial volume fraction  $\Phi$  of the suspension and found a linear law which we were able to explain with a model taking into account the balance between the volume energy and the repulsive energy between aggregates, without introducing a surface energy. In the following section of this paper we propose a model including both the repulsive energy between the aggregates and the surface energy which predicts the average size of the aggregates by a minimization of the total magnetic energy analogous to the one used for the determination of the deformation of ferrofluids droplets [8-10]. In section 3, we present some experimental results and compare them with the theoretical predictions. We conclude by a discussion of the validity of this model and of its application to electrorheological suspensions.

## 2. Theoretical model.

We consider the case of a magnetic suspension submitted to an external magnetic field  $H_0$  much higher than the field corresponding to the transition and we model the structure of the suspension by a hexagonal array of ellipsoids of transverse semi-axis  $b$  and of length  $L$  (cf. Fig. 1). In the dipolar approximation the ratio  $\lambda$  of the magnetic energy to

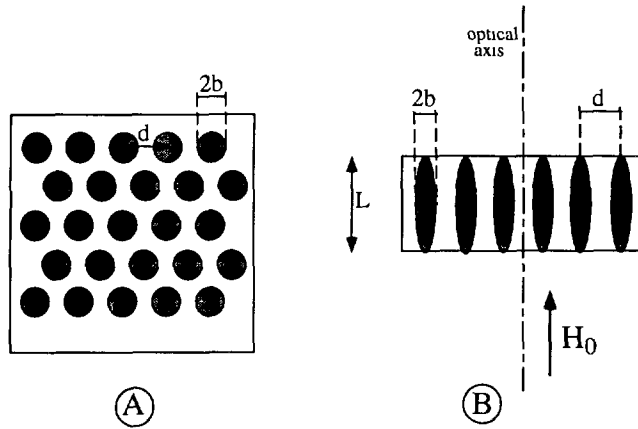


Fig. 1. — Schematic diagram of the structure A : top view B : side view. The aggregates are assimilated to prolate spheroids of semi-axes  $b$  and  $\ell$ .  $L = 2 \ell$  is the thickness of the cell.

$kT$  is given by  $\lambda = (4 \pi \mu_0) \mu_1 \beta^2 H^2 / (4 kT)$  where  $\mu_1 = 1$  is the magnetic permeability of the liquid phase. In our experimental case we find  $\lambda = 2.63 H^2 (\text{Oe})$ . For the terminal field we have used (50 Oersted) we obtain  $\lambda = 6\,500$ . Owing to this high value of  $\lambda$  we can neglect the thermodynamic forces and suppose that the internal structure of the aggregates is the B.C.T. equilibrium structure with a density  $\Phi_a = 2 \pi / 9$ . This is also only under these conditions that we can use an equation similar to (1) which assumes that the surface energy is localized on the last layer of particles. On the other hand the magnetic field is still low enough so that we can consider that the magnetic permeability  $\mu$  is almost constant, then the magnetic energy is given by [11] :

$$U_T = - \frac{N_a \mathbf{M}_a \cdot \mathbf{H}_0}{2} \tag{2}$$

with  $N_a$  the number of aggregates and  $\mathbf{M}_a$  the magnetic moment of an aggregate. If we consider a spheroidal (prolate) aggregate (cf. Fig. 1) of semi-axes  $b$  and  $\ell$ , with  $b/\ell < 1$ , composed of an homogeneous magnetic medium of average permeability  $\mu_a$ , the internal magnetic field is constant inside the aggregate and the homogeneous magnetic moment is given by (in e.m.u.)

$$\mathbf{M}_a^h = \frac{\mu_a - 1}{4 \pi} \left[ \frac{\mathbf{H}_0}{1 + n_z (\mu_a - 1)} \right] V_a \tag{3}$$

where  $V_a$  is the volume of the aggregate and  $n_z$  the demagnetizing factor which for an ellipsoid of excentricity  $e = (1 - b^2/\ell^2)^{1/2}$  is given by

$$n_z = \left[ \frac{(1 - e^2)}{2 e^3} \right] \left[ \ln \left( \frac{1 + e}{1 - e} \right) - 2 e \right]. \tag{4}$$

If the ratio of the two axes of the ellipsoid  $\kappa = 2 b/L$  is very small, we have  $n_z \sim -4 (1 + \log \kappa) \kappa^2$ , and minimizing the surface energy (Eq. (1)) plus the volume energy (Eqs. (2) and (3)) with respect to  $b$  gives the behavior  $b \propto a (L/a)^{2/3}$  reported in [7]. Let us see more precisely the physical mechanism.

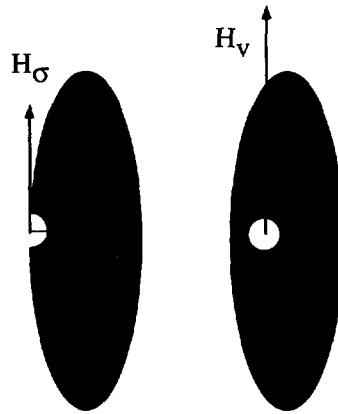


Fig. 2. — The local field,  $H_\sigma$ , on a spherical particle located on the surface is lower than the local field,  $H_v$ , on a particle in the bulk.

As the ellipsoid elongates the demagnetizing factor tends to zero, then the magnetic moment and the absolute value of the energy increase continuously so we could expect that the average radius,  $b$ , of the aggregate will tend to the radius of a particle. Actually we have to take into account two other energies which, on the contrary are favorable to the growth of  $b$ . One is the repulsive energy,  $U_r$ , between the aggregates and has been discussed in a previous paper [1], and the other,  $U_\sigma$  is a correction to the volume energy calculated on the basis of a continuous medium, coming from the finite size of the particles. This contribution can be understood as a surface energy which can be calculated numerically and depends on the configuration of the first two or three layers of particles below the surface [12]. In order to understand the origin of this energy and to simply estimate its value in a mean field approach, let us compare the local magnetic field on a spherical particle inside the aggregate  $H_v$  to the local field  $H_\sigma$  on a particle located on the surface (cf. Fig. 2). For a homogeneous ellipsoid whose main axis is aligned with the field in the  $z$  direction we have

$$\mathbf{H}_v = \mathbf{H}_0 + \frac{4\pi\mathbf{P}}{3} - 4\pi n_z \mathbf{P}.$$

The field  $4\pi\mathbf{P}/3$  where  $\mathbf{P} = \mathbf{M}_a^h/V_a$  comes from the inner surface formed by the spherical cavity which represents the exclusion volume of a particle (Fig. 2) and  $4\pi n_z \mathbf{P}$  is the demagnetizing field coming from the external surface of the ellipsoid. For a particle on the surface, if we assume that the polarization  $\mathbf{P}$  remains constant inside the ellipsoid, the difference with the former case will come from the absence of polarization on a half spherical surface which will give for the surface field :

$$\mathbf{H}_\sigma \approx \mathbf{H}_0 + \frac{2\pi\mathbf{P}}{3} - 4\pi n_z \mathbf{P}.$$

The local field on the particles situated on the surface is then lower by a factor  $2\pi\mathbf{P}/3$  relatively to the bulk local field. Then the real magnetic moment of the aggregate,  $\mathbf{M}_a$ , will be lower than that,  $\mathbf{M}_a^h$ , calculated for a homogeneous ellipsoid :

$$\mathbf{M}_a = \mathbf{M}_a^h - N_1 \delta \mathbf{m}_1 \quad (5)$$

where  $\delta \mathbf{m}_1 = \alpha^p 2 \pi \mathbf{P}/3$  is the difference between the dipolar moment of a particle inside the aggregate and the one on the surface;  $\alpha^p = a^3 \beta$ , with  $\beta = (\mu_p - 1)/(\mu_p + 2)$ , is the polarizability of a spherical particle.  $N_1$  is the number of particles on the surface of the ellipsoid. With this cavity model  $\delta \mathbf{m}_1$  is positive and corresponds to a surface tension since it decreases the total dipole of the aggregate and so increases the electrostatic energy. Actually the value of  $\delta \mathbf{m}_1$  is quite sensitive to the precise structure on the surface. With the notation of [12] we have more generally  $\delta \mathbf{m}_\nu = \mathbf{m}(M_\nu - 1)$  where  $\mathbf{m}$  is the bulk dipole of a particle inside the aggregate and  $M_\nu = m_\nu/m$  the normalized dipole of the layer  $\nu$  ( $\nu = 1$  being the surface layer). Using for the bulk dipole  $\mathbf{m} = \mathbf{M}_a^h/N_p$  where  $N_p$  is the number of particles inside the aggregate we can generalize (5) by writing :

$$\mathbf{M}_a = \mathbf{M}_a^h(1 - n_\sigma) \quad \text{with} \quad n_\sigma = \frac{1}{N_p} \sum_\nu N_\nu (\tilde{M}_\nu - 1). \quad (6)$$

In the homogeneous cavity model only the surface layer is concerned and we have  $(M_1 - 1) = (\alpha^p 2 \pi \mathbf{P}/3)/m$ , or using  $P = N_a m/V_a$  and  $\alpha^p = \beta a^3$ , we obtain  $(M_1 - 1) = \beta \Phi_a/2$ .

In a lattice model we have shown in [12] that the contribution of the surface layer was far greater than all the others, so we can still keep only the first layer. For a B.C.C. lattice and  $\mu_p/\mu \rightarrow \infty$  an exact calculus gives  $(M_1 - 1) = 0.3$  for  $\Phi = 0.65$  and  $(M_1 - 1) = 0.37$  for  $\Phi = 0.67$ . We did not calculate this quantity for a B.C.T. lattice but we do not think that the result would be very different. In any way this result is not far from  $\beta \Phi_a/2$  and we shall keep this value as a good estimate of the relative change of dipole moment on the surface. We have still to estimate  $N_1$  the number of particles on the surface,  $S_a = 2 \pi b^2 + 2 \pi (b\ell/e) \sin^{-1} e$ , of the ellipsoidal aggregate. If we consider a surface with a hexagonal packing we find  $N_1 = S_a/(2 \sqrt{3} a^2)$ . This packing corresponds to the denser planes of the B.C.T. lattice and we shall take it as the upper realistic limit of  $N_1$ . With these values of  $N_1$  and of  $(M_1 - 1)$  we obtain

$$n_\sigma = a \frac{S_a}{V_a} \beta \frac{\pi}{3 \sqrt{3}}. \quad (7)$$

If we introduce the expression (6) of  $\mathbf{M}_a$  in the total magnetic energy (cf. Eq. (2)), the term with  $n_\sigma$  will represent the contribution of the surface energy  $U_\sigma$ . On the other hand, we also need to consider the repulsive interactions between the aggregates. If we assume a structure formed by a two-dimensional hexagonal array of ellipsoidal aggregates, each one bearing the same total moment  $\mathbf{M}_a$ , we can find the moment  $\mathbf{M}_a^h$  of a given aggregate,  $i$ , considered as a homogeneous medium, by writing

$$\mathbf{M}_a^h = \alpha^a \left( H_0 - \sum_j \mathbf{T}_{ij}^r \mathbf{M}_a \right). \quad (8)$$

The quantity  $\mathbf{T}_{ij}^r$  is the propagator of the field between two aggregates,  $i$  and  $j$ , of length  $L$  whose axes of symmetry are separated by a distance  $d_{ij}$ . It can be obtained easily in the dipolar approximation, with all the polarization located on the revolution axis of each aggregate. In this case the field of one aggregate is equivalent to the field  $\mathbf{H} = \sigma \mathbf{r}/r^3$  of two charges  $\sigma^\pm = \pm M_a/L$  located on each extremity of the axis of revolution of the ellipsoid. Because of the cylindrical symmetry we can consider only the  $z$  component of the field in the sum over all the aggregates. Then the average  $\langle H \rangle_i = -\mathbf{T}_{ij}^r \cdot \mathbf{M}_a$  over the length of the

aggregate  $i$  of the field coming from the aggregate  $j$  defines the quantity  $T_{ij}^r$  which will be given by

$$T_{ij}^r = \frac{1}{L^2} \left[ \frac{2}{d_{ij}} - \frac{2}{(d_{ij}^2 + L^2)^{1/2}} \right]. \quad (9)$$

Of course, if  $d_{ij}/L \gg 1$ , we recover the dipolar propagator for parallel dipoles in the same plane  $T_{ij}^r = 1/d_{ij}^3$ .

The polarizability,  $\alpha^a$  corresponds to the ratio  $M_a^h/H_0$  for an isolated aggregate and is given by equation (3). The total magnetic energy per unit volume is still given by equation (2) but with now  $M_a$  obtained from the solution of equations (6) and (8). We thus obtain

$$\frac{U_T}{V} = - \frac{\Phi H_0^2}{\Phi_a 8 \pi} \left[ \frac{(\mu_a - 1)}{1 + n_z(\mu_a - 1)} \right] \left[ \frac{(1 - n_\sigma)}{1 + n_r(1 - n_\sigma)} \right]. \quad (10)$$

The ratio  $\Phi/\Phi_a$  comes from the equality  $N_a V_a/V = \Phi/\Phi_a$ . The values of  $n_z$  and  $n_\sigma$  are given by equations (4) and (6) respectively and  $n_r = \alpha^a \sum_j T_{ij}^r$  represents the effect of the repulsive interaction of one aggregate with all the others. The value of  $n_r$  can be calculated by summing (9) on a triangular lattice or by replacing the sum by an integral : the two results are within 2 % so we shall take the analytical result which is

$$n_r = \left[ \frac{(\mu_a - 1)}{1 + n_z(\mu_a - 1)} \right] \left[ \frac{\Phi}{\Phi_a} \right] \left[ \sqrt{\left(1 + \frac{d_0^2}{L^2}\right)} - \frac{d_0}{L} \right] \quad \text{with} \quad d_0 = b \sqrt{\frac{2 \Phi_a}{3 \Phi}} \quad (11)$$

The quantity  $d_0$  is the radius of a cylindrical cavity containing the reference aggregate and surrounded by a homogeneous medium of average density  $\Phi$  instead of the hexagonal distribution of figure 1. It is worth noting that the use of equations (6) and (8) amounts to introduce in a self consistent way the surface energy of an aggregate and the repulsive interaction with the other aggregates.

### 3. Experimental results and comparison with the model.

The experiments have been performed with a suspension of magnetic particles at two volume fractions  $\Phi = 10 \%$  and  $\Phi = 5 \%$ . The particles are spherical but polydisperse with an average diameter of  $0.8 \mu\text{m}$ . They are composed of the same grains of magnetite which usually compose a ferrofluid but, instead of being free in the liquid phase, these grains are imprisoned in a polystyrene matrix. The size of the grains is less than  $100 \text{ \AA}$  and even if these grains do not rotate in the matrix their magnetic moment can jump from one direction of easy magnetization to another by thermal activation since we have  $KV_g \ll kT$  where  $K$  is the anisotropy constant and  $V_g$  the volume of the grains of magnetite. By measuring the magnetization curve on a dried powder we have verified that these particles were superparamagnetic and that we had no hysteresis. The magnetic permeability,  $\mu_p$ , of the particles has been deduced from the measurements of the average magnetization of the suspension at several volume fractions with the help of Bruggeman's theory [13]. For a magnetic field of a few tens of Oersteds, we obtain  $\mu_p = 37$ . Note that the permeability was constant in this low field domain. The suspension was placed between two transparent disks whose distance,  $L$ , is set by different spacers from  $150 \mu\text{m}$  to  $700 \mu\text{m}$ . The field is perpendicular to the disks and is raised very slowly to its terminal value (50 Oersted) in order to get an equilibrium structure. The typical rising time was 0.1 Oersted per minute and we have verified that for longer times the results were the same.

The terminal value of 50 Oersted corresponds to the completion of the aggregation process as observed by light transmission : above this value there was no further increase of the transmitted light [1].

The structure is observed with a microscope whose optical axis is parallel to the field. The images are recorded on a computer and treated with an image analysis in order to obtain the position of the center of gravity of each aggregate. A typical view of the projection of the aggregates on a plane perpendicular to the field is represented in figure 3A for  $\Phi = 5\%$  and in figure 3B for  $\Phi = 10\%$ . At the highest volume fraction the projections do not correspond to an ellipsoidal shape for the aggregates. This branched shape has some similarity with the patterns observed in the ferrofluids when the initially ellipsoidal droplet bends and distorts in a complicated pattern which depends on the intensity and on the rate of increase of the magnetic field [14]. The dependence of the shapes of the aggregates on the final amplitude of the field

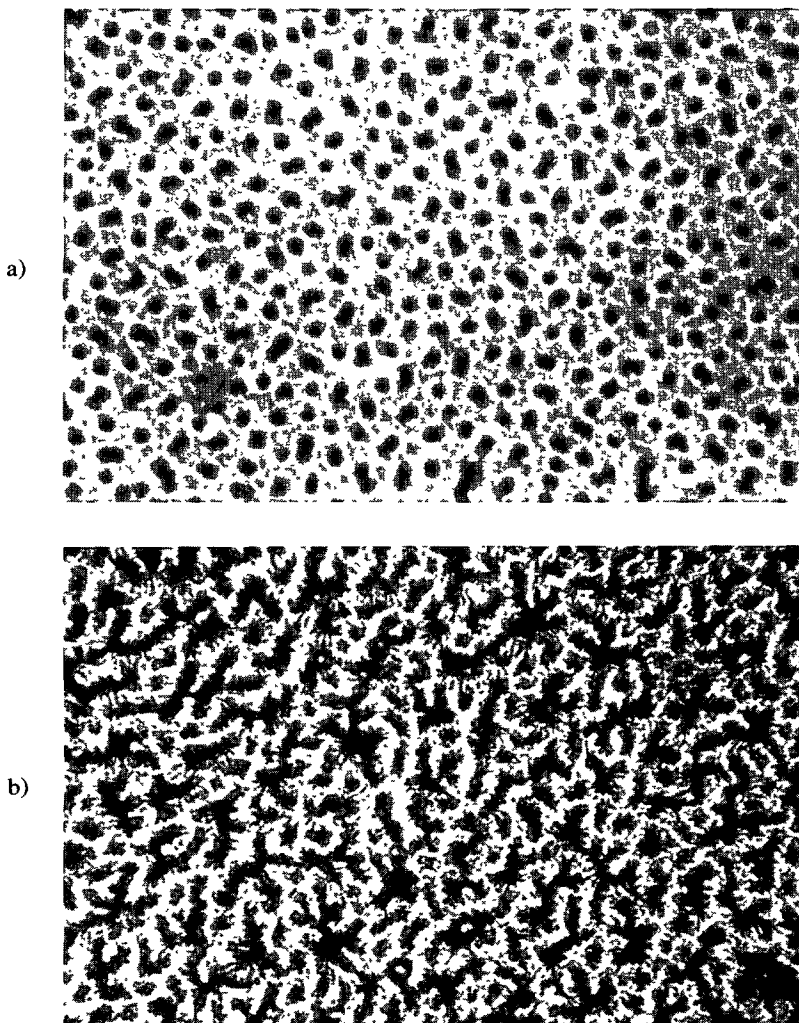


Fig. 3. — real structure observed by optical microscopy. The view corresponds to a projection on a plane perpendicular to the magnetic field (which is along the axis of observation) 3a : Volume fraction of 5 % ; 3b : Volume fraction of 10 %.

and on the time history of its application has also been observed recently on micronic droplets of ferrofluids [15]. On the other hand, on another device where the upper disk can translate [19], we can experimentally see that all the aggregates connect the two walls of the cell.

We can measure experimentally by image analysis the average distance,  $d$ , between the centers of gravity of the aggregates. This is possible as long as these aggregates remain well individualized even if their projections are not simple disks (cf. Fig. 3). We obtain  $d$  by taking the maximum of the pair correlation function  $g(r)$  of the separations between the aggregates [1]. In order to relate this experimental distance to the semi-axis  $b$  of our idealized ellipsoidal aggregate we represent the structured suspension as shown in figure 1. In this situation we have

$$b = d \sqrt{\frac{3\sqrt{3}\phi}{4\pi\phi_a}} \quad (12)$$

where  $\phi_a = 0.69$  is the volume fraction inside the aggregates.

The log-log plot of the experimental values of  $b$ , versus the thickness of the cell, is represented in figure 4 (solid circles) for a volume fraction of 10 % and in figure 5 (same symbols) for a volume fraction of 5 %. These values can be fitted by a straight line with a slope of  $0.67 \pm 0.04$  and of  $0.68 \pm 0.04$  respectively. On the same figures are plotted the values obtained by the minimization of the total magnetic energy (cf. Eq. (10) with respect to  $b$  for the same values of  $L$  (empty circles). We see that the predictions are 40 % too low for the volume fraction of 10 % but are very close to the experimental values for 5 %. The discrepancy at  $\Phi = 10\%$  is not so unexpected in view of the non-ellipsoidal shape of the aggregates. We also have to emphasize that the internal permeability of the aggregates is directly obtained from Bruggeman's theory : (with  $\mu_p = 37$  and  $\phi_a = 0.69$  we obtain  $\mu_a = 22$ ) so we have no free parameter in this model.

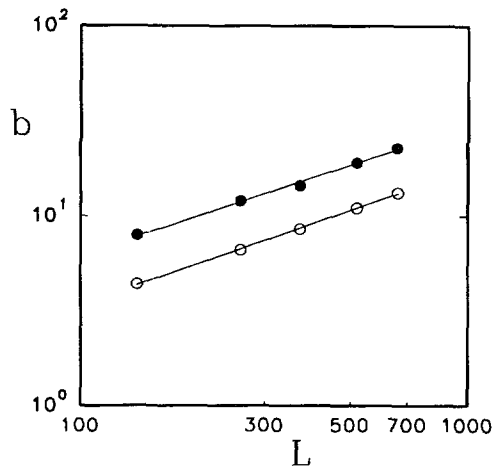


Fig. 4. — Log-Log plot of the transverse semi-axis of the aggregates versus the thickness of the sample for a magnetic suspension of initial volume fraction  $\Phi = 10\%$ . (○) Theoretical prediction from equation (10) (Slope 0.71). (●) Experimental results (Slope  $0.67 \pm 0.04$ ) obtained by image analysis with the help of equation (12) to relate  $b$  and  $d$ .



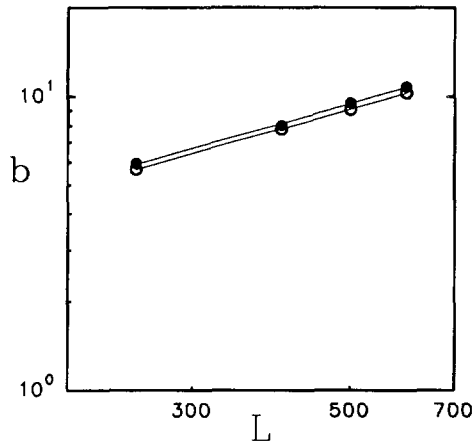


Fig. 5. — Same as for figure 4 with a volume fraction :  $\Phi = 5\%$ . (○) Theoretical prediction from equation (10) (Slope 0.68). (●) Experimental results obtained by image analysis (Slope  $0.68 \pm 0.04$ ).

It appears that this model is quite efficient in predicting the average size of the aggregates if the volume fraction is not too high. We have calculated for the same magnetic suspension at a volume fraction of 5 % the size of the aggregates for a much larger range of cell thicknesses :  $10 < L < 10\,000\ \mu\text{m}$ . The results are presented in figure 6 (solid curve) on a log-log plot. We first notice that we do not have a straight line : there is no well defined power law defined for the whole range of distances. For low values of  $L$  :  $10\ \mu\text{m} < L < 50\ \mu\text{m}$  we find an exponent of 0.35, for the intermediate range where we have done some experiments, we find an exponent close to  $2/3$  and in the range  $10\,000\ \mu\text{m} < L < 20\,000\ \mu\text{m}$  we find 0.85. Ultimately when the effect of the surface tension becomes totally negligible the exponent will tend towards unity since in this case all the terms in the energy only depend on the ratio  $b/L$ . This is the case for the dotted curve in figure 6 which represents the prediction of the model without the surface energy ( $n_\sigma = 0$ ). The other limit where we neglect the repulsive

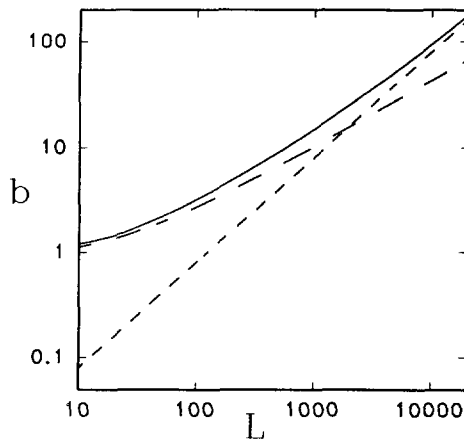


Fig. 6. — Theoretical prediction of the transverse semi-axis,  $b$ , of the aggregates versus the thickness  $L$  of the sample for a volume fraction of 5 %. Full theory : equation (10). Equation (10) with  $n_\sigma = 0$  : no surface tension. Equation (10) with  $n_r = 0$  : no repulsion between aggregates.

energy ( $n_r = 0$ ) and only take into account the surface energy is represented by the dash-dotted curve. For large thicknesses we tend to an exponent  $2/3$ ; in the range  $100 \mu\text{m}$ - $700 \mu\text{m}$  we have a slope of 0.57 and at the beginning of the curve a slope of 0.33. Note that, with the value of the surface tension given by [7], the surface effect is dominant at low thicknesses and in our experimental range the values are only 20-30 % too low. This would not appear if we take the dipolar approximation for the surface energy equation (1) which gives much lower values of  $b$ .

Other experiments with a larger range of thicknesses would be useful to verify the predictions of the model and we are designing a new sealed cell which operate in a larger range. Some similar experiments have also been carried out recently by Liu *et al.* [15] on a suspension of rigid ferrofluid droplets. They have explored the range of thicknesses between 10 and  $500 \mu\text{m}$  and found the same qualitative behavior as we predict: there is no constant exponent for the variation of  $b$  with  $L$  and at low thicknesses we tend to an exponent close to  $1/3$ .

#### 4. Conclusion.

We have shown that it was possible to predict the equilibrium structure induced by a magnetic field in a magnetic suspension if we introduce both the repulsive interactions between the aggregates and the surface energy due to the finite size of the particles. This study was performed with a magnetic suspension mainly because we have precise measurements of the magnetic permeability which furthermore is much easier to obtain than the permittivity of a suspension (in the latter case, problems are met due to the ionic clouds surrounding the particles and to the charge on the electrodes). We have made some preliminary experiments with an electrorheological suspension composed of silica particles activated by water in a low viscosity silicone oil. The cell was formed of two glass disks coated with transparent electrodes. Unfortunately our particles slightly aggregate under the effect of electrostatic forces: they disintegrate under mixing but when the field is turned off, the Brownian motion itself does not allow to recover a homogeneous state. In these conditions we have some irreversible aggregation and the model based on a minimization of the electrostatic energy alone will probably fail. On the other hand the aggregates were not so well defined and the uncertainty on the distance was larger. Nevertheless we have tried some preliminary experiments on this suspension and we have found that in the same range of thicknesses  $100 < L < 700 \mu\text{m}$  the size of the aggregates was increasing with an exponent close to 0.6. As already said in the introduction the electrostatic case is different because the charges brought by the electrodes cancel the depolarizing field of each aggregate. Then, at first sight, the model should apply if we set the repulsive interaction to zero ( $n_r = 0$  in Eq. (9)) and indeed the exponent of 0.6 is quite compatible with a surface tension effect (cf. Fig. 6). Nevertheless experiments with particles which do not stick together or on the electrodes should be made in order to get equilibrium structures in the presence of an electric field.

A systematic investigation with different particle sizes, different permittivities (which can be varied by changing the frequency of the applied field), should enable us to decide if this model allows the size of the structures in electro and magnetorheological fluids to be predicted or if it is restricted to some set of parameters. The prediction of the structure induced in quiescent suspensions by an external field is the first step in the study of a broader class of ordering phenomena, in particular, shear induced ordering which is found in complex fluids like micellar fluids, nematics, suspensions [16, 17] and electrorheological suspensions. In these last ones we have observed the formation of stripes of particles aligned parallel or perpendicular to the flow [18, 19] in the presence of to an electric field and of a sinusoidal shear. Actually, these E.R. or M.R. fluids are very convenient for the study of shear induced

ordering since we can very easily modify the energy of interactions between the solid particles by changing the applied field. Such systems should help to the elaboration of a theory able to explain shear induced ordering.

### Acknowledgments.

This work was realized with the financial support of Rhône Poulenc. We are also very grateful to A. Audoly and B. Gay-Para for their technical support.

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