J. Phys.: Condens. Matter **2** (1990) SA291–SA294. Printed in the UK

Magnetic lyotropic phases

J C Dabadie†, P Fabre†, M Veyssié†, V Cabuil‡ and R Massart‡

† Collège de France, Laboratoire de Physique de la Matière Condensée, 11, Place Marcelin Berthelot, 75231 Paris Cédex 05, France

‡ Université Pierre et Marie Curie, Laboratoire de Physico-Chimie Inorganique, 75252 Paris Cédex 05, France

Received 9 July 1990

Abstract. We have demonstrated that it is possible to include tiny magnetic particles into different types of lyotropic phases, such as sponge, microemulsion or lamellar phases. The first point of interest in these results is to prove the compatibility between solid colloids and organized liquids.

As for the hybrid lamellar phase, we have studied its phase diagram versus the smectic period and the particle concentration—which are the two relevant parameters—and deduced its range of stability. Moreover, this ferrosmectic phase exhibits original features when subjected to a magnetic field, even when it is very low: the lamellae orientate in the direction of the field. The detailed mechanism of this strong coupling between the spherical particles, the flexible membranes and the magnetic field is not fully understood, and deserves further experimental and theoretical study.

1. Introduction

In the framework of an investigation of the eventual compatibility of colloidal particles and structured liquid phases [1, 2], we have studied hybrid systems associating a ferrofluid suspension with different lyotropic mesophases. A reasonable probability of succeeding was expected as swollen lyotropic phases have recently been obtained [3, 4], whose characteristic size is of the same order of magnitude as colloidal particles. In this article, we present recent results which prove the existence of several stable ferrophases, and describe some effects of a magnetic field on them.

2. Magnetic mesophases

We start with a quaternary mixture, consisting of oil (cyclohexane), water, surfactant (sodium dodecyl sulphate) and cosurfactant (pentanol). The phase diagram of this system has been studied by di Meglio *et al* [5] at room temperature. The main interesting features, for our purpose, are the following:

(i) existence of lamellar phases, which may be swollen by oil while keeping constant the thickness of the water layer, in a wide range of size from 4 to 40 nm.

0953-8984/90/SA0291 + 04 $03.50

(ii) Existence of two isotropic phases, adjacent to the lamellar domain; one of them is a 'sponge' phase, characterized by a bicontinuous topology [6]; the other one is a water-in-oil microemulsion.

It is important to emphasize that one may go from the sponge phase to the lamellar one and, ultimately, to the microemulsion just by increasing the cosurfactant (alcohol) content. We have prepared these different phases and checked them by small angle x-ray scattering.

On the other hand, Cabuil and Massart have prepared a colloidal suspension of magnetic particles, following the method described in [7]: microspheres of ferrimagnetic material (γFe_2O_3) are dispersed in oil (cyclohexane) and stabilized by adsorbed organo-phosphorated molecules. The average diameter of the particles, of the order of 10 nm, and their size distribution are determined by analysis of the magnetization curve [8] and x-ray technique [2]. The particle volume fraction of this ferrofluid may be varied from 0.1% to 10%.

Using then the ferrofluidic lipidic solution instead of pure cyclohexane, we attempt to rebuild the lyotropic phases previously described. We observe that stable homogeneous phases incorporating magnetic particles may be obtained with sponge, lamellar and microemulsion structures. The stability of these different phases depends both on the particle concentration c and on the characteristic size e of the undoped lyotropic system. In fact, it appears that these two parameters do not act independently, but we may distinguish some general trends as follows:

(i) At very low concentration ($c = 0.2\%$), the doped phase is more stable than the initial ferrofluidic solution.

(ii) On the other hand, when increasing the magnetic concentration in the lamellar structure, the stability is improved for thinner layers.

These two unexpected features are probably due to magnetic interactions, which seem to stabilize quasi 2D systems more efficiently than the isotropic ones. This problem deserves more experimental work, such as a parallel study with non-magnetic solid particles, as well as theoretical reflection.

3. Magnetic field effects in lamellar phases

Usual ferrofluids are very sensitive to magnetic fields [9], exhibiting several types of instabilities, due to their pronounced superparamagnetic susceptibility. In contrast, smectic phases usually do not respond to magnetic fields, except at very high values, because of the conjunction of a poor diamagnetic anisotropy and a large elastic rigidity [10].

In the present hybrid system, we have observed a very specific behaviour for the lamellar phases [1]: anisotropic field effects occur at very low field values (of the order of 20 G). When the magnetic field is applied to an orientated lamellar sample, nothing happens if the field is parallel to the lamellae; if the field is perpendicular to the lamellae, defects appear (figure 1(a)) for a threshold value H_c of the field; when H is increased beyond H_c, the defects grow in number and finally merge into a typical fan-shaped texture [10] all over the sample (figure 1(b)). This process clearly indicates a reorientation of the layers in the field direction, which starts with a nucleation of defects possessing a toric symmetry.

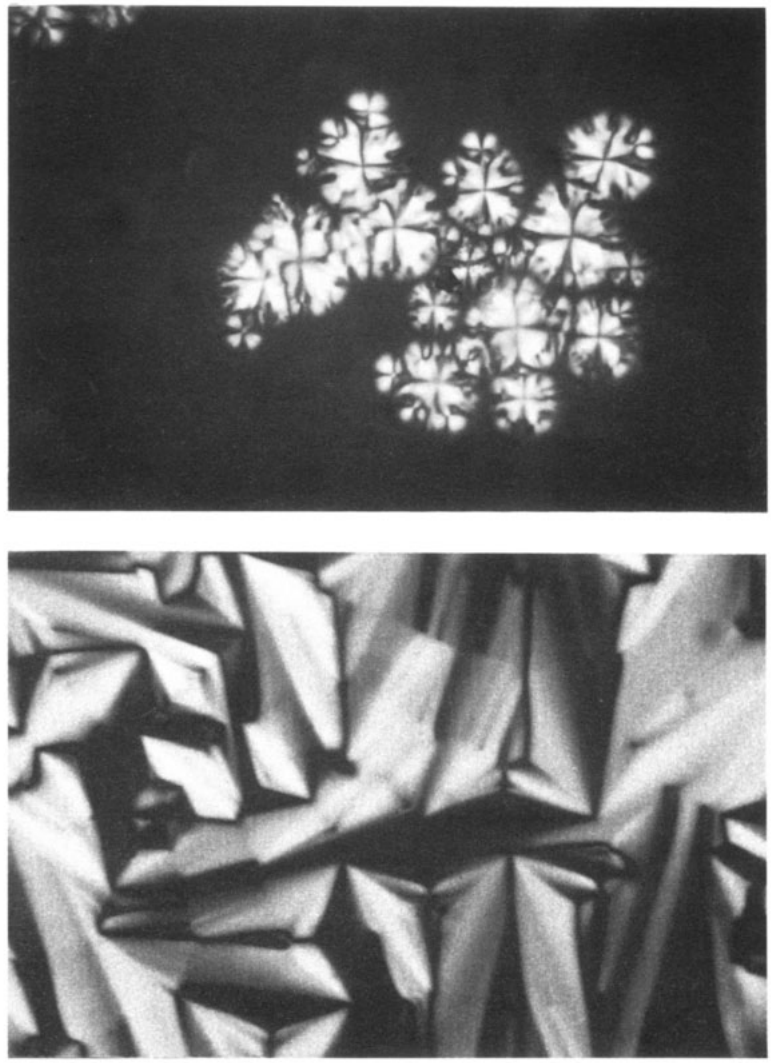

Figure 1. Magnetic field effects on lamellar phases; (*a*) isolated toric defects nucleation; (*b*) evolution towards a fan-shaped texture.

We have measured the H_c value as a function of the pertinent parameters, mainly the thickness of the sample d and the magnetic concentration c; we find that H_c scales as $d^{-1/2}c^{-1/2}$ (figure 2). The dependency on the sample thickness is usual for the field effects in liquid crystals, and has been predicted for the smectic case in different models [11]. The variation with the magnetic concentration is more puzzling. First of all, there is no standard theory describing the magnetic anisotropy of such a material; we had previously developed a simple model [1] where the layers were treated as infinite and independent slabs of magnetic medium; a classical demagnetizing field calculation then leads to a c^{-1} law for H_c, in clear disagreement with our recent experimental results; improvements of the model, taking into account the finite size and the layer's coupling

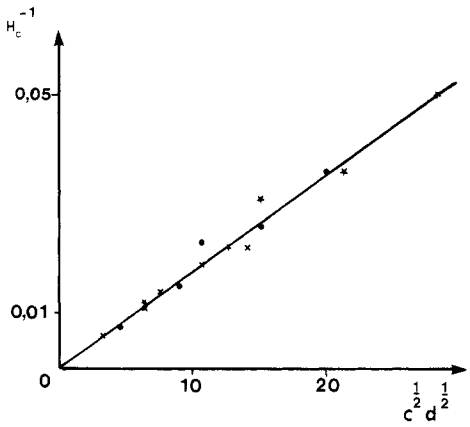

Figure 2. A plot of the inverse magnetic field threshold, $1/H_c$, as a function of $d^{-1/2}c^{-1/2}$, where d is the sample thickness and c the magnetic concentration.

effect, do not modify the value of the exponent. We then have to search for a much more anisotropic model, for instance in terms of magnetic dumbells confined in 2D layers.

4. Conclusion

The existence of these new magnetic phases opens a wide domain of theoretical reflection and experimental investigations: one has to make clear the effects of 2D magnetic interactions on the stability of these structures, but also on the dynamical properties of the lamellae when incorporating solid particles. Light scattering studies are currently underway, and we expect that they will lead to precise and local information. More generally, it will be interesting to study equivalent systems, with non-magnetic solid microparticles incorporated in organized fluids, including the limiting case of a unique bilayer membrane.

References

[1] Fabre P, Casagrande C, Veyssié M, Cabuil V and Massart R 1990 *Phys. Rev. Lett.* **64** 539
[2] Fabre P, Ober R and Veyssié M 1990 *J. Magn. Magn. Mater.* **85** 77
[3] Larche F C, Appell J, Porte G, Basereau P and Marignan J 1986 *Phys. Rev. Lett.* **56** 1700
[4] Safinya C R, Roux D, Smith G S, Sinha S C, Dimon P, Clark N A and Bellocq A M 1986 *Phys. Rev. Lett.* **57** 2718
[5] di Meglio J M, Dvolaitzky M and Taupin C 1987 *Physics of Complex and Supermolecular Fluids* ed S A Safran and N Clark (New York: Wiley) p 361
[6] Gazeau D, Bellocq A M, Roux D and Zemb T 1989 *Europhys. Lett.* **9** 447
[7] Massart R 1981 *IEEE Trans. Magn.* **MAG-17** 1245
[8] Bacri J C, Perzynski R, Salin D, Cabuil V and Massart R 1986 *J. Magn. Magn. Mater.* **62** 36
[9] Rosensweig R E 1985 *Ferrohydrodynamics* (New York: Cambridge University Press)
[10] de Gennes P G 1974 *The Physics of Liquid Crystals* (Oxford: Clarendon)
[11] Parodi O 1972 *Solid State Commun.* **11** 1503