

Phase Transitions in Soft Condensed Matter

Edited by
Tormod Riste and
David Sherrington

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Phase Transitions in Soft Condensed Matter

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Series B: Physics

Phase Transitions in Soft Condensed Matter

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PREFACE

This volume comprises the proceedings of a NATO Advanced Study Institute held in Geilo, Norway, between 4 - 14 April 1989. This Institute was the tenth in a series held at Geilo on the subject of phase transitions. It was the first to be concerned with the growing area of soft condensed matter, which is neither ordinary solids nor ordinary liquids, but somewhere in between.

The Institute brought together many lecturers, students and active researchers in the field from a wide range of NATO and some non-NATO countries, with financial support principally from the NATO Scientific Affairs Division but also from Institutt for energiteknikk, the Norwegian Research Council for Science and the Humanities (NAVF), The Nordic Institute for Theoretical Atomic Physics (NORDITA), the Norwegian Physical Society and VISTA, a research cooperation between the Norwegian Academy of Science and Letters and Den norske stats oljeselskap a.s (STATOIL).

The organizing committee would like to thank all these contributors for their help in promoting an exciting and rewarding meeting, and in doing so are confident that they echo the appreciation also of all the participants.

Soft condensed matter is characterized by weak interactions between polyatomic constituents, by important thermal fluctuations effects, by mechanical softness and by a rich range of behaviours. The main emphasis at this Institute was on the fundamental collective physics, but preparation techniques and industrial applications were also considered.

The introductory lectures set the scene and introduced the nature of the constituent systems and their cooperative consequences and conceptual make-ups for the whole field, with later talks emphasizing sub-fields in greater detail. The lectures were supplemented by research seminars and poster sessions, all of which proved conducive of considerable discussion and continuing exchange. These proceedings include write-ups of the majority of the lectures and seminars and a selection of the posters.

One important class of soft condensed matter comprises thermotropic liquid crystals, which are composed of relatively small and simple molecular building blocks but produces a plethora of interesting phases characterized by orientational and spatial order. These are the subject of several articles.

Polymers are composed of larger molecules. Many aspects of their phase transitions, association and gelation were discussed and reported here, as also other physical properties.

Amphiphilic molecules are soap molecules with a polar head and an alkyl tail, able to bind respectively to polar liquids and hydrocarbons. Structures formed by amphiphilic molecules in contact with water are two-dimensional films and membranes, which may further interact to form lyotropic liquid crystals with a wide range of polymorphism and interesting vibrational and defect properties, which were discussed, as also were lipid mono- and bi-layers and Langmuir-Blodgett films. In water-oil mixtures, surfactant amphiphilic molecules, binding to both constituents, lead to microemulsions and to micelles, of great interest to the petroleum and food industry, as well as possessing fascinating properties to excite the physicist.

Many interesting aspects are associated with the static and dynamic properties of liquid surfaces and interfaces and with the wetting of solid surfaces.

Fractals have been a subject of considerable interest in recent years and were the subject of the Institute in 1985. An ideal system to study their statics and dynamics is that of areogels and this study was a natural link between this Institute and the 1985 Institute. Other aspects of fractals and growth were also reported.

As noted earlier, although the emphasis was on fundamental microscopic physics, industrial applications were also discussed and illustrated: soft condensed matter is already a major part of our lives and its use is growing. An interesting application involving modern polymeric processing to illustrate fundamental features is the use of perfectly engineered polystyrene spheres and related shapes immersed in water or ferrofluid to model microparticle interactions on a scale providing ready optical observations. In a sense this study (second lecture) represents a sort of "periodic boundary conditions" on the Institute, but in fact the subject is growing in many conceptual dimensions, as the discussions reported here amply demonstrate.

The subject of phase transitions in soft condensed matter is still in its infancy, compared with more conventional condensed matter physics, but it possesses a richness and a potential which are likely to provide excitement and challenges for many years to come. We hope that the papers presented in this volume will help germinate in others the euphoria which is already rapidly growing in the participants of the tenth Geilo Institute.

Finally, we would like to express our deep gratitude, and that of ten generations of participants, to Gerd Jarrett of the Institutt for energiteknikk, Kjeller, Norway who did all the practical organization of all ten of the Geilo Institutes, including the preparation of their proceedings, with incredible efficiency and smoothness. Without Gerd the series would not have been the success it seems to have been.

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Adsorption of polymers has a broad spectrum of industrial applications as a tool to control the stabilization of colloidal suspensions[1]. The problem is to overcome the aggregation tendency of solid particles, due to their van der Waals attraction and one idea is to sterically protect the colloid with polymers[2]. The steric protection is usually achieved by adsorbing or grafting polymers onto the colloidal particle. If the colloid is much bigger than the polymer one can model the colloidal particle by a flat surface. Adsorption from a good solvent, which swells the chains, leads to a fluffy layer with a thickness of order of R_G , the radius of gyration of the chain in the solvent[3,4]. Grafting the chains by one end at high densities actually forces the chains in a highly stretched conformation and the layer size becomes proportional to N , the polymerization index of the chains[5,6].

The adsorbed layer is well described by a self-similar picture[4] generated by a geometrical constraint on the screening length ξ of the semi-dilute solution near the wall: the only distance in the problem being the distance z from the wall one must have $\xi = z$ which leads to a concentration profile

$$\phi(z) \simeq \left(\frac{a}{z}\right)^{\frac{4}{3}} \quad (1)$$

The grafted layer can also be viewed as a semi-dilute solution[5] but the correlation length ξ is now equal to the distance D between the anchoring points. If a blob of size D has g monomers, one chain has N/g blobs, the layer thickness is then

$$L = \frac{N}{g} D = Na\sigma^{\frac{1}{3}} \quad (2)$$

where $\sigma = (a/D)^2$ is the density of anchoring points and a the size of a monomer.

In addition to homopolymers a large variety of other architectures can be used to achieve colloidal stabilization. We investigate here the interfacial behavior of diblock and random copolymers. Diblock copolymers adsorb[7,8] in a bi-layer structure which depends on several physico-chemical parameters like the surface affinity, the solvent quality or the mutual incompatibility of the blocks. We discuss next the adsorption from a selective and a non-selective solvent.

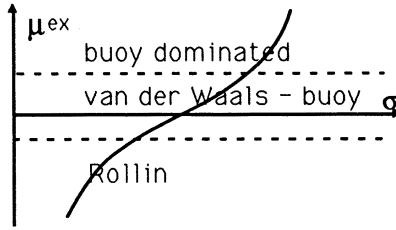


Figure 1. Variation of the chemical potential μ_{ex} as a function of the surface density σ

1. Selective solvent.[7] In this case the solvent is poor for one of the blocks and good for the other. An isolated chain in the bulk would have thus a collapsed block — let's say of N_A monomers of type A (the anchor) — and a swollen block of N_B monomers of type B (the buoy). The collapsed block sticks on the wall in order to avoid contact with the solvent. It forms, in the limit of an infinite selectivity of the solvent, a molten layer of A monomers. This layer anchors the swollen blocks that point towards the solvent building-up a grafted layer.

Diblock copolymers are known to form mesophases[9,10] in selective solvents: micelles, lamellae, worm-like micelles ... These aggregation effects are relevant for the interfacial behavior of the copolymers: the bulk solution acts as a reservoir for the adsorbed layer and imposes the chemical potential μ_{ex} . To study the adsorbed layer we write the surface grand canonical free energy of the layer as

$$\Omega = F_A + F_B - \mu_{ex}\sigma + \Pi_{ex}(L_A + L_B) \quad (3)$$

where F_A and F_B are the contributions of the anchor A and of the buoy B layers, L_A and L_B their respective thicknesses and Π_{ex} the sum of the external pressures acting on the layer: osmotic, mechanical... The anchor free energy has three different contributions measured in units of $k_B T$:

$$F_A = -S + \frac{3L_A^2\sigma}{2N_A} + \frac{A}{6\pi L_A^2} \quad (4)$$

S is the spreading power that takes in account the various surface tensions[11]. The second term on the right-hand side of equation (4) is the contribution of the stretching energy of the chains in the molten layer[12]. The third term accounts for the direct interactions between the wall-anchor and anchor-solvent interfaces[13], which we approach by a van der Waals energy. A is there the Hamacker constant which is positive in this case: the interaction tends to pull the surfaces apart. In a melt there is no confinement energy of the chains[14].

The buoy is in a grafted layer configuration and has a surface free energy[5]:

$$F_B = N_B\sigma^{\frac{11}{6}} \quad (5)$$

Since the A blocks form a molten layer

$$N_A a = \frac{L_A}{\sigma} \quad (6)$$

L_B is also a function of σ given by Eq.(2) and the minimization of the grand canonical potential leads to the equilibrium curve $\sigma = \sigma(\mu_{ex})$ sketched in figure 1. Different regimes should be distinguished for the configuration of the adsorbed layer according to the value of the chemical potential:

- (a) *Rollin regime* When the solution is very dilute the chemical potential is strongly negative. At equilibrium the chemical potential term balances the van der Waals energy

$$L_A \sim a \left(\frac{\mu_{ex}}{N_A} \right)^{-1/3} ; \quad L_B \sim \frac{N_B}{N_A^{2/9}} \mu_{ex}^{-1/9} \quad (7)$$

- (b) *van der Waals-Buoy regime* In the concentration range where micelles form in the bulk, the chemical potential is [7]

$$\mu_{ex} \simeq N_A^{2/5} \gamma_{AS}^{3/5} \quad (8)$$

For a small asymmetry ($\beta = (R_B/R_A) \simeq 1$) the chemical potential is positive and drives the chain towards the layer. But the insertion of extra chains in the layer increases the stretching energy. The balance between these two terms leads to the equilibrium sizes

$$L_A \sim a N_A^{12/25} \beta^{-2} \sim R_A ; \quad L_B \sim R_B N_A^{4/25} \simeq R_{micelles} \quad (9)$$

- (c) *Buoy dominated regime* If micelles are present in the bulk and the asymmetry of the copolymer is large ($\beta \gg 1$) the anchoring layer has a thickness smaller than the radius of the chains in the melt and the chains in the buoy are almost extended:

$$L_A \sim a N_A^{6/23} \beta^{-10/23} \ll R_A ; \quad L_B \sim N_B^{21/23} N_A^{-12/23} \gg R_{micelles} \quad (10)$$

2. Non-selective solvent.[8] In this case the solvent is good for both blocks and one of the blocks adsorbs preferentially on the surface, the other one is strongly repelled by the surface. The layer can be modelled by a self-similar continuous layer anchoring an external grafted layer. As above we define an asymmetry ratio $\beta = (R_B/R_A)$. The free energy has the same form as in the previous case — Eq.(3) — but the contribution from the anchor has a functional dependence on the concentration profile $\phi_A(z)$. Thus one needs to minimize the grand canonical potential with respect to $\phi_A(z)$ and σ , obeying the internal constraint of connection of blocks *A* and *B*

$$\sigma = \frac{1}{N_A} \int_0^\infty dz \phi_A(z) \quad (10)$$

The profile of the *A* monomers is given by Eq.(1) but it extends only over a distance L_A smaller than R_G which depends on β . For asymmetry ratios smaller than $N_A^{1/2}$ the characteristics of the layer are

$$\sigma = \frac{1}{N_A}; \quad L_A \sim \frac{N_A^{1/2}}{\beta} a; \quad L_B \sim N_B N_A^{-1/3} \quad (11)$$

For larger asymmetries the thickness of the adsorbed layer becomes of order of a monomer size and the surface density is β dependent

$$\sigma \sim \beta^{-2} \quad (12)$$

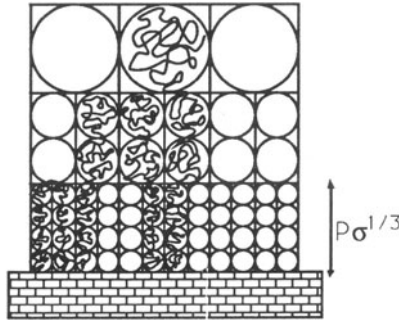


Figure 2. Blob picture for a random chain adsorbed from a dilute solution. Near the wall the chains are stretched. On the outer side the chains build a self-similar layer.

When the asymmetry is very large ($\beta \gg N_A^{3/4}$) the anchor breaks into small individual pancakes but the chains in the buoy remain in a brush configuration.

3. Random copolymers.[15] The copolymer considered in this paragraph is built up from a long repulsive chain of monomers where a fraction f of stickers A has been introduced randomly. Random copolymers often adsorb in an intermediate configuration between the grafted layer made by block copolymers and the adsorbed layer of homopolymers.

We first study the adsorption of an isolated chain : for an idealized (gaussian) isolated chain the adsorption profile extends much further in the solvent than the equivalent adsorbing chain of A monomers. The diagrammatic expansion of a perturbation serie can be resummed to study in detail the behavior of the chain. More qualitatively it is possible to give a simple Flory argument balancing the loss of entropy due to confinement with the adsorption energy gained by contact with the surface. In the case of a penetrable interface this reads

$$\left. \frac{F}{T} \right|_{chain} = -\frac{\delta}{P} \varphi M + \left(\frac{a}{D} \right)^2 M \quad (13)$$

where P is the average number of monomers B between two adjacent stickers A . In the first term, on the right hand side, $\varphi = a/D$ is the fraction of monomers in contact with the surface, a fraction $1/P$ of those (the monomers A) being able to gain an energy $-\delta$. The second term is the loss of entropy of a gaussian chain of M monomers confined in a size D . Minimizing this energy with respect to D leads to

$$D \sim \frac{aP}{\delta} \quad (14)$$

The adsorbed random copolymer thus has loops extending f^{-1} times further in the solvent than a simple homopolymer. For a solid wall there is a depletion layer close to the wall, decreasing the number of B monomers in contact with the interface by an additional P factor. This leads to a confinement length

$$D \sim \frac{aP^2}{\delta} \quad (14)$$

with loops extending even further in the solvent.

In the more realistic case where the chains are adsorbed from a solution of finite concentration we propose a blob model — figure 2 — to describe the layer. If the density of attached stickers is higher than $P^{-\frac{6}{5}}$ the chains in the vicinity of the surface are stretched over a distance of order Pa . For larger distances the layer crosses over smoothly from a grafted to an adsorbed configuration. The outer part of the layer has thus a self-similar profile.

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