Food Dispersion Stability

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1 Introduction

As a world's leading food processor, Kraft Foods is deeply interested in gaining a fundamental understanding of the stability of its food products, many of which are food emulsions and/or foams. Emulsions and foams are thermodynamically unstable and will separate over time. The primary modes of destabilization of emulsions involve creaming, flocculation and coalescence. These processes occur concurrently and tend to build on each other. In the case of foams, coalescence and Ostwald ripening are the primary culprits. While we admit that Ostwald ripening has not been seriously investigated as much as other mechanisms, we believe it is not very prevalent in food systems.

In the case of emulsions, Hartland and Gakis[1] and later Lobo *et al.*[2] developed models of stability of batch emulsions which tend to cream. Van den Tempel[3] and Borwankar *et al.*[4] have accounted for simultaneous flocculation and coalescence using simple models incorporating 'average' flocculation and coalescence rate constants and other simplifying assumptions. We later developed[5] a more detailed model by extending Smoulchowski theory to account for coalescence within aggregates. At the core of all these attempts to describe more rigorously the stability of emulsions is the coalescence rate constant. Over the past three or four decades, it has been generally accepted that coalescence in foams and emulsions is controlled by the thinning and rupture of thin liquid films between bubbles bubbles or droplets. Hence, as have focused the majority of our effort on the role of thin films in the stability of food emulsions and foams.

This paper summarizes the salient findings of our most recent work in the area of food dispersion stability. Stratification and location of emulsifier play a major role in governing the drainage and stability of thin films, especially in the

case of low-molecular-weight surfactants. Particularly interesting are the results on non-equilibrium systems which are able to rationalize the otherwise contradictory results between theory and practice. Proteins, the major workhorse in the stabilization of food emulsions and forms, make a fascinating, albeit complicated, study. Through the study of thin films of proteins we are now beginning to unravel the details of protein stabilization of emulsions and foams. While much of our attention has been focused on the study of single thin liquid films and their role in governing food dispersion stability, we have also recently started investigating emulsion systems in a macroscopic way using Kossell diffraction, which has allowed us to study the role of fat particle structure in the stabilization of foamed emulsions. In this paper we first present the microscopic world of thin liquid films and later move on to reporting the investigations using Kossell diffraction. We hope to show that we have pushed the limits of the application of the techniques for thin film studies and Kossell diffraction from model systems to real systems. Results on real food systems will be presented.

2 Thin Film Studies

The first thin film studies revolved around the use of hydrodynamic modelling to determine the lifetimes of thin liquid films as they drain and eventually rupture. These models highlight the roles of interfacial tension gradients (Gibbs–Marangoni effect) and interfacial viscoelasticities (see, for example, references 6 and 7). But, in relation to food dispersions, these models have three major shortcomings. Firstly, the interfacial tension gradient mechanism fails to explain the stability of emulsions which are prepared by incorporating an emulsifier in the droplet phase, since according to this mechanism locating the emulsifier in the droplet phase should be detrimental to the interfacial tension gradients which are needed to be sustained for emulsion stability. Secondly, the mechanism fails to explain the stability behaviour of emulsions prepared at surfactant concentrations above the critical micelle concentration (CMC), where interfacial tension gradients are diminished. And, thirdly, these models predict lifetimes of thin films to be of the order of seconds or at most minutes, whereas food dispersions are required to be, and are, stable for days, weeks or months. Later, a mechanism of stratification or micellar stabilization was observed[8] which addresses the second point. Eventually, this mechanism was extended[9] to show that thermodynamically metastable films can result under certain conditions, thereby addressing the third point.

In our investigations of systems of interest to food industrial practice, we have discovered new mechanisms relevant to low-molecular-weight surfactants under non-equilibrium conditions which provide satisfactory explanations of practical observations. We have also now firmly established that structure formation, closely related to stratification, is widely prevalent in food systems and is likely to be responsible for their long term stability.

Emulsion Stabilization under Non-equilibrium Conditions

Low-molecular-weight surfactants which are soluble both in water and in oil are frequently used as emulsifiers. Normally, they are initially dissolved in one of the phases only. Just after formulation of the emulsion, there is a time period when intensive surfactant redistribution and mass transfer through the interfaces takes place. On the other hand, during this period the adsorption of amphiphilic molecules at the newly created oil–water surfaces may still be incomplete. When two droplets with such unsaturated and highly mobile oil–water interfaces collide, it is clear that, due to the lack of sufficient surface elasticity and viscosity, the thin liquid film between them will drain very fast, and rupture will eventually occur, unless other factors favouring stabilization are present. Since some time has to be allowed until a tightly packed rigid adsorption layer of surfactant develops, whereupon direct repulsive forces (steric, electrostatic, structural) come into play and so providing long-term stability, our attention is directed to the initial transit period which may be crucial in determining whether the emulsion will survive. Our model studies with thin liquid films made in a glass capillary have shown that new stabilizing effects are operative in non-equilibrium conditions, when the surfactant is being transferred from one of the bulk phases to the other. We have looked at both cases, one where the surfactant is initially dissolved in the droplet phase and the other where it is initially dissolved in the thin phase.

At pointed out by Hartland,[10] when a surface-active solute diffuses across the interface from the continuous phase (film) to the dispersed phase (droplet), this should lead to stabilization. Because the volume of the film is small, the solute concentration there falls more rapidly than in the bulk of the continuous phase, and hence the interfacial tension is greater around the film centre and correspondingly smaller at the periphery and in the meniscus. A tensile restoring force emerges, which is opposite in direction to the surfactant density gradient[11] (*i.e.*, from higher to lower surface pressure). It sets the fluid interface in motion, and liquid is dragged towards the film centre. This is a manifestation of the well-known Marangoni effect,[10,11] and is expected to provide high stability. Such a qualitative picture has been confirmed by recent experiments,[12] although it turns out that the real processes are far more complex.

In the paper of Velev *et al.*,[12] a fascinating cyclic phenomenon was encountered during observations of aqueous emulsion films between oil phases in the presence of non-ionic surfactant (soluble both in water and in oil). When the surfactant is initially dissolved in the film phase, and diffuses across the interface towards the oil, its transfer induces spontaneous growth of a lens-like region (dimple) around the film centre (Figure 1). The thickness along the periphery remains more or less constant. Upon reaching a certain size, the dimple flows out into the meniscus, and a new one starts to form. The process is cyclic and usually goes on for many hours. Its driving force was proven to be the surfactant mass transfer.[12] A clear distinction must be made between this cyclic dimpling and the so-called hydrodynamic dimple. The latter appears only when the film is thinning, and is not connected in any respect with mass transfer through the

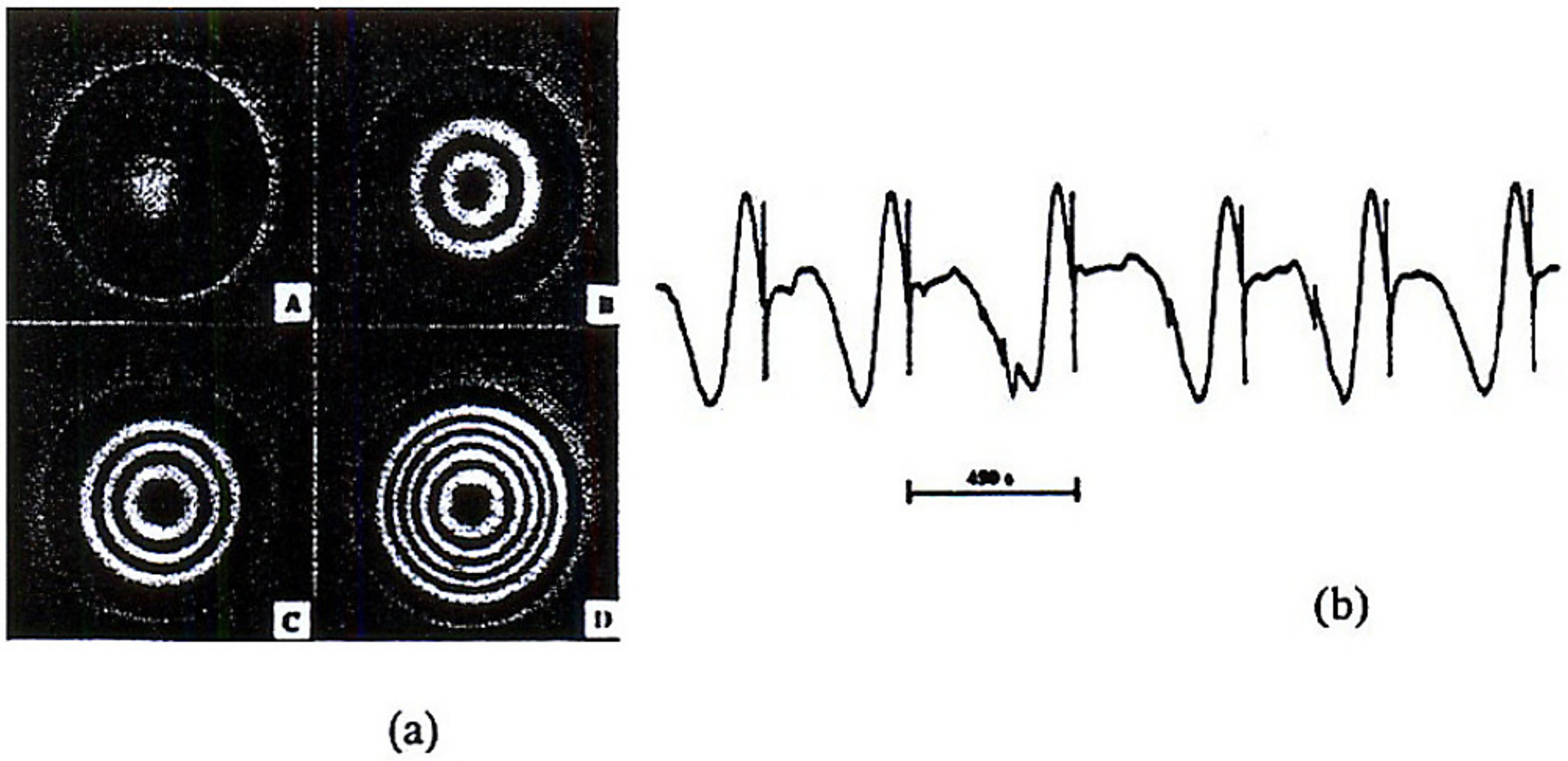


Figure 1 *Oscillating dimple in an emulsion film (diameter ≈ 330 μm). (a) Interferometric image in reflected light of wavelength λ. Black and white colours correspond to multiples of λ/(4n), where* n *is the refractive index in the film; (b) intensity as a function of time of light reflected from a small fixed area of the film.*

phase boundaries. The film surfaces are deformed under the action of the viscous friction. At a certain stage of thinning the hydrodynamic dimple irreversibly disappears and the film becomes plane-parallel.

A comprehensive theoretical description of the process of dimple growth has been proposed by Danov *et al.*[13] These authors have proven that the molecular surface diffusion and the surface viscosity are insignificant factors in the phenomenon. Moreover, the diffusion flux of surfactant from the film phase towards the interface turns out to be negligible as well. The main flux is by surface convection, *i.e.*, by the inward flow along the oil–water boundary, directed from the meniscus to the film, which provides a continuous supply of new surfactant (see Figure 2). A major role can be attributed to the Marangoni effect, connected with interfacial tension gradients. As a fluid element moves to the film centre, along the interface, it continuously loses surfactant because the latter goes to the oil phase. Therefore, the surface concentration decreases and the interfacial tension σ rises with diminishing values of the radial coordinate r. The induced gradients of σ set the fluid surface into motion. Thus, liquid is dragged into the film, feeding the dimple, as shown in Figure 2.

The theoretical calculations of dimple shapes are in good agreement with experimental dimple profiles measured interferometrically.[13] Quasi-stationary shapes are possible for not very big diffusion fluxes (j_0). Otherwise, large negative pressures develop near the film periphery and eventually lead to expulsion of the dimple. The analysis of the flow properties reveals the appearance of vortices inside the film; they show up after a certain period of dimple growth. In this system the stabilizing effect, which keeps the film very thick for a long time, can be attributed to hydrodynamic reasons, in particular,

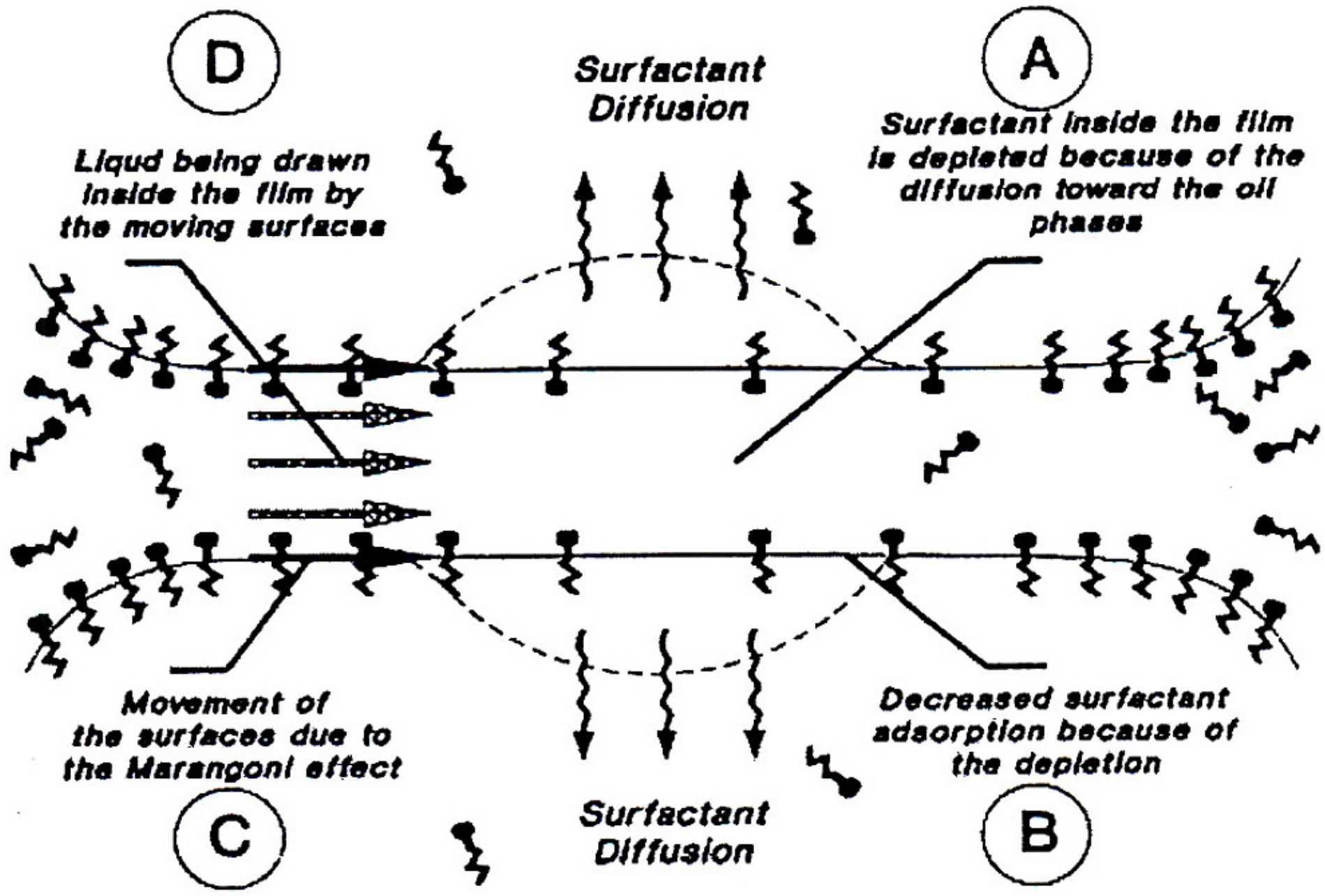


Figure 2 *Schematic illustration of how the surfactant fluxes lead to the phenomenon of the oscillating dimple.*

to net repulsion when the pressure is integrated along the liquid–liquid interfaces.

The complementary case, when surfactant is initially dissolved in the oil (droplets) and afterwards diffuses across the phase boundaries towards the aqueous film, is even more interesting.[14] Very good stability was observed, contrary to the expectations based on the conventional theory, which maintain that locating emulsifier in the droplet phase is detrimental to formation of interfacial tension gradients which must be sustained to confer emulsion stability.[6] One should also point out that this stability cannot be attributed to electrostatic or any other conventional type of repulsion. Experiments were carried out[14] with aqueous films between xylene phases, in the presence of non-ionic surfactant (Tween 20) which was initially dissolved in the oil. The water contained 0.1 mol l^{-1} NaCl. The films remained thick (above 100 nm) and absolutely stable, until the equilibrium distribution of surfactant was reached (which took up to 48 hours). Then, the films thinned down and ruptured. In this system one observes a specific dynamic pattern, consisting of intensive liquid circulation through channels and exchange of mass in the lateral direction between the film and the Plateau border (see Figure 3).

In order to clarify the physical mechanism of the phenomenon, the surfactant concentration in the oil phase was varied.[14] It turns out that the effects emerge in a threshold manner: below a certain 'critical' concentration, there is no stabilization at all. Experiments were performed also with films whose phases contained given amounts of surfactant (C_0) before being put in contact with the oil. The stabilizing effect and the general pattern of the process were found to be

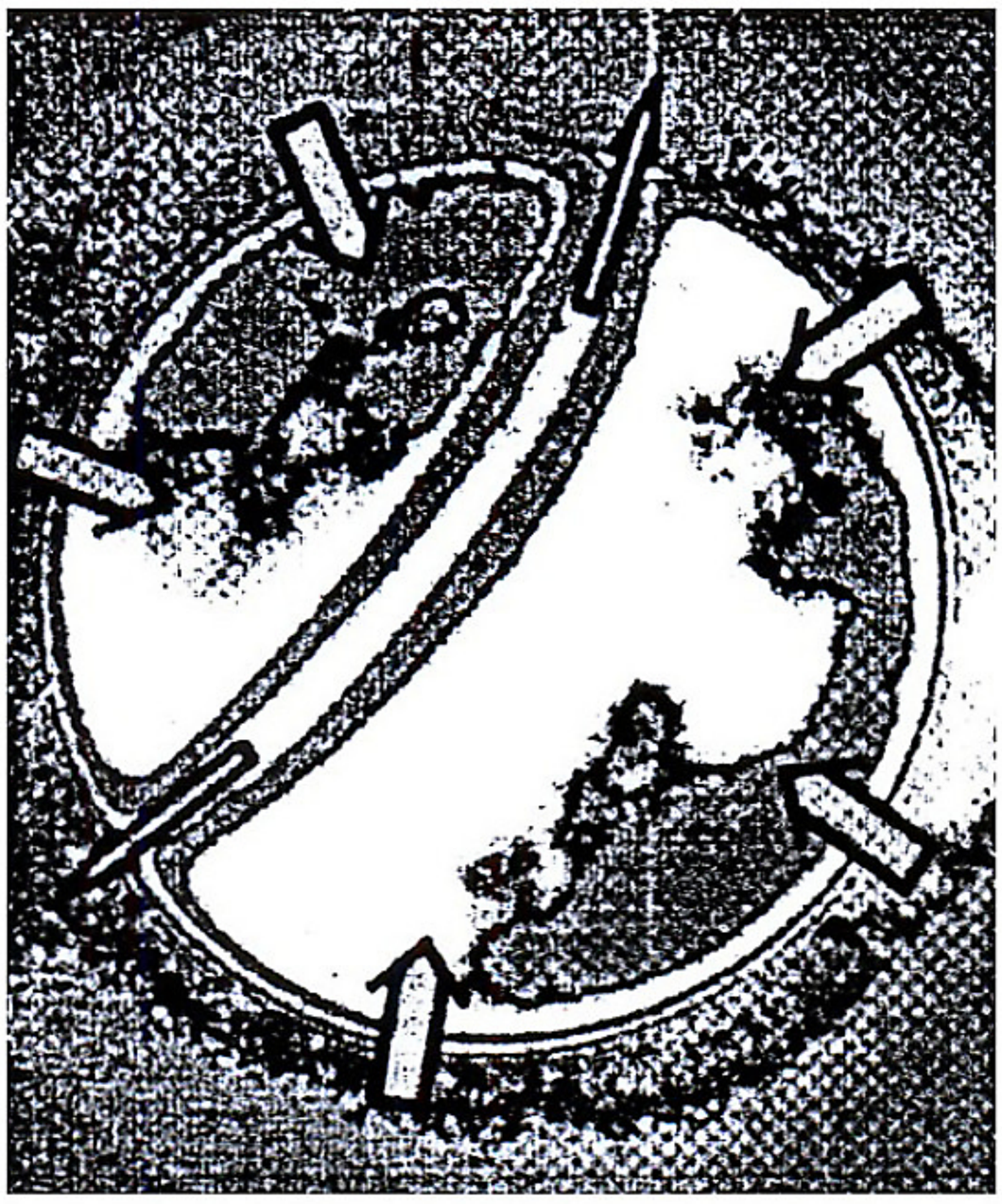


Figure 3 *Aqueous film stabilized by 5 × 10^{-4} M Tween 20. The surfactant is initially dissolved in the surrounding xylene phase and diffuses to the film across the interfaces. The arrows indicate liquid fluxes brought about by gradients in osmotic pressure of surfactant micelles.*

the same for $C_0 = 0, 1, 10, 100$ and even 500 × CMC. The value C_0 represents a background concentration, evenly distributed both in the film and in the meniscus during the experiment (Figure 4). Such a high surfactant content does not influence the film stability, which indicates that only the *excess* concentration in the film with respect to the meniscus is important.

These findings may be rationalized by the following explanation (see Figure 4). As the surfactant diffuses through the interfaces, its concentration in the aqueous film gradually increases. Two types of solute species exist in the water phase: single molecules (monomers) and micellar aggregates. The diffusion of the monomers is fast, and they are uniformly distributed in the aqueous phase. The local monomer concentration in the film cannot increase above the CMC because the individual molecules, after crossing the oil–water boundary, quickly form micelles. As the monomer concentration in water is constant, the diffusion flux coming from the oil cannot stop until the equilibrium distribution is reached. The thin and broad film gets enriched with micelles, whose concentration in the voluminous Plateau border remains low (the Brownian diffusion of the big aggregates is slow). The increased osmotic pressure of *micelles* in the film engenders convective fluxes of liquid; material is sucked from the meniscus, and after saturation with surfactant it rushes out through the channels of higher thickness (Figure 3).

As discussed by Ford *et al.*,[15] the primary role of emulsifiers is stabilizing emulsions that arise upon droplet rupture during high energy emulsification in

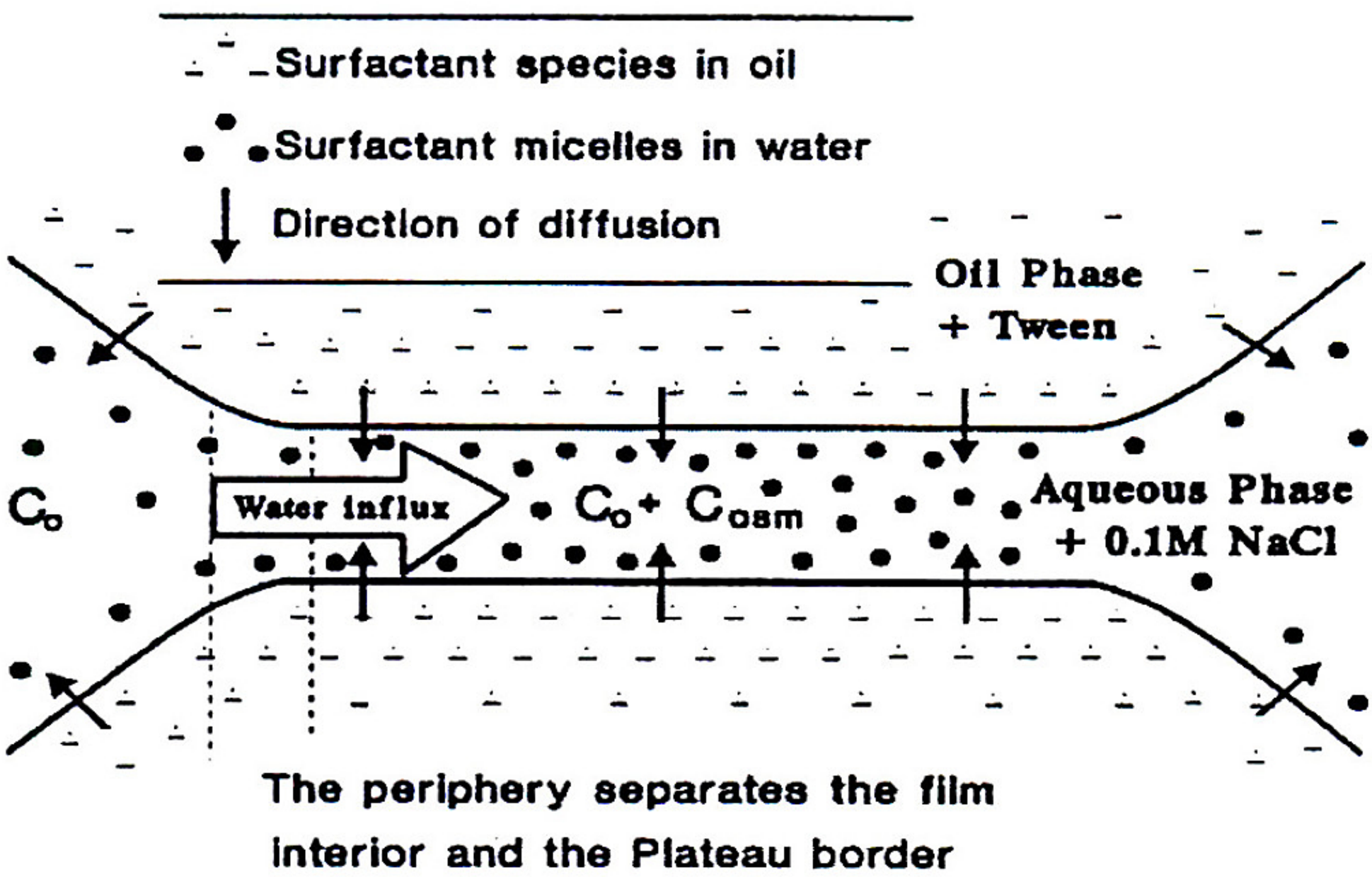


Figure 4 *Sketch of an aqueous film explaining the origin of osmotic pressure differences and liquid fluxes which are due to the non-equilibrium distribution of surfactant micelles.*

conventional emulsion processing equipment. 'Fast' emulsifiers adsorb rapidly so that the interfacial tension of interest in droplet breakage is quite close to the equilibrium interfacial tension, resulting in what Armbruster calls mechanically limited dispersion.[16] Then it was not very apparent to us how such systems may be stabilized under the intense environment during emulsification (high compressive forces during collisions). Perhaps the mechanisms discussed above are relevant. For a 'slow' emulsifier (Armbruster's example is egg yolk), the operative interfacial tension during breakage is much higher than the equilibrium interfacial tension, resulting in mechanically limited dispersion. In addition, the resulting emulsion is not stabilized by the above mechanisms because no significant equilibration of egg yolk occurs, since its proteins are not soluble in the oil phase, and recoalescence is not completely prevented.

Stratification in Food Systems

The conventional (DLVO) theory for the stability of dispersions takes into account only two kinds of molecular interactions: electrostatic and van der Waals. An important type of non-DLVO surface force is the structural interaction, which manifests itself when the continuous phase contains small colloidal particles such as, for example, micelles or latex spheres. In general, whenever a surface bounds a liquid phase, ordering is induced among the particles close to the wall. In the case of a film, the structured regions near the two opposing surfaces overlap; this gives rise to an oscillatory disjoining pressure and interaction energy.[9] This effect causes a step-wise thinning of the

Table 1 *Effect of protein concentration on the number of step transitions of a foam or emulsion film formed from sodium caseinate at 40 °C or above*

Concentration (wt%)	*Number of step transitions*
0.01	0
0.1	1
0.5	2
2	3
4	4

thin liquid film (so-called 'stratification'), recognized as the layer-by-layer destruction of the colloidal crystal of spherical particles inside the film.

Koczo *et al.*[17] studied thin film systems of interest for food emulsions and foams approach using the interferometric technique. They observed a layering of caseinate sub-micelles in thin films made with sodium caseinate solution which was similar to the stratification observed with surfactant micelles and latex particles. As shown in Table 1, the number of step transitions was found to increase with increasing concentration of sodium caseinate, and for the extent of stratification it did not matter whether the film was a foam film or an emulsion film (with oil phase of *h*-hexadecane). The calculated effective volume fraction of the sub-micelles in a 2 wt% solution of caseinate is 20%. The three-step transitions observed at this concentration are in good agreement with previous work[9] in which three-step transitions were observed for non-ionic and anionic micellar systems in the range of 10–20% effective volume fraction.

The layering of sodium caseinate sub-micelles in foam and emulsion films results in increased drainage time as the layers are removed one-by-one via a series of step transitions. Furthermore, under certain conditions (low temperature, small film size), the film transition can be completely inhibited so that drainage stops with the film still containing one or more layers of micelles. Such films are rather thick and so can be very stable. Thus, the layering of sub-micelles can prevent two oil droplets or air bubbles from approaching together. This is proposed as a new mechanism of stabilization for these systems.

Such layering need not be confined specifically to sodium caseinate. Indeed, Koczo *et al.* proposed[17] that layering could occur in other systems, especially those containing globular proteins. One of the systems of interest to us is similar to a frozen whipped topping formulation. Frozen whipped toppings are prepared by making an oil-in-water emulsion followed by cooling to crystallize the fat. The emulsion is then aerated and whipped to create a whipped topping that can be frozen. Typically, sodium caseinate and lipid emulsifiers are used along with gums. Our investigations have revealed that polysorbate micelles, and even macromolecules of the food hydrocolloids studied (xanthan and guar gum), can induce layering in thin liquid films. And it was found that, when sodium caseinate, lipid emulsifiers and gums are all present simultaneously, as

Table 2 *Effect of various food ingredients on the number of film transitions of a foam film*

Solution	*Temperature (°C)*	*Number of step transitions*
0.01% Polysorbate 60	25–80	0
0.1% Polysorbate 60	65, 80	1
0.5% Polysorbate 60	45	0 (film size < 0.3 mm)
0.5% Polysorbate 60	45	2 (film size > 0.3 mm)
0.5% Polysorbate 60	65, 80	2
0.5% Polysorbate 60 + 2% sodium caseinate	25, 65	3
0.5% Polysorbate 60 + 2% sodium caseinate	80	4
Polysorbate 60, sodium caseinate, xanthan, guar, sugars, corn syrup	65–80	6–9
0.1% xanthan gum		a
0.05% guar gum		a

[a] Step transitions observed, but number not recorded; step height equals size of random coil of gum molecule, indicating microlayering of gum molecules.

in a real whipped topping system, up to 9 film transitions could be observed (see Table 2).[18]

Model Investigations on Emulsion Systems Stabilized by Proteins

The amphiphilic nature of proteins makes them suitable for stabilizing oil-in-water emulsions, especially in food applications. Here we discuss some recent results from model studies with two types of proteins: globular (BSA, bovine serum albumin; BLG, β-lactoglobulin) and disordered (β-casein). Their molecular structures have been characterized in bulk aqueous solutions, *e.g.*, circular dichroism and hydrodynamics, but not yet fully determined for molecules adsorbed on liquid interfaces.[19]

Marinova *et al.*[20] have explored the properties of thin aqueous films (diameter 200–400 μm) using a Scheludko cell. This approach has been shown to provide insights regarding the interaction of two approaching protein-stabilized interfaces under a variety of conditions of ionic strength, pH, specific ion effects, and ageing time. Specifically, the technique has been used to measure the interaction energies between pairs of interfaces through their contact angles as well as their film thickness. With the Scheludko cell, electrostatic interactions have been shown to be operative at pH values away from the isoelectric point of the protein. It has been found[20] that, without added salt, the films containing 0.015 wt% BSA at pH = 6.6 (isoelectric point pI = 4.8) remain very thick and stable. Even a minor amount of inorganic electrolyte (as little as 10^{-3} M NaCl) is sufficient to screen the electrostatic repulsion, and this results in the formation of very thin Newton black films. Another piece of evidence[20] for the importance

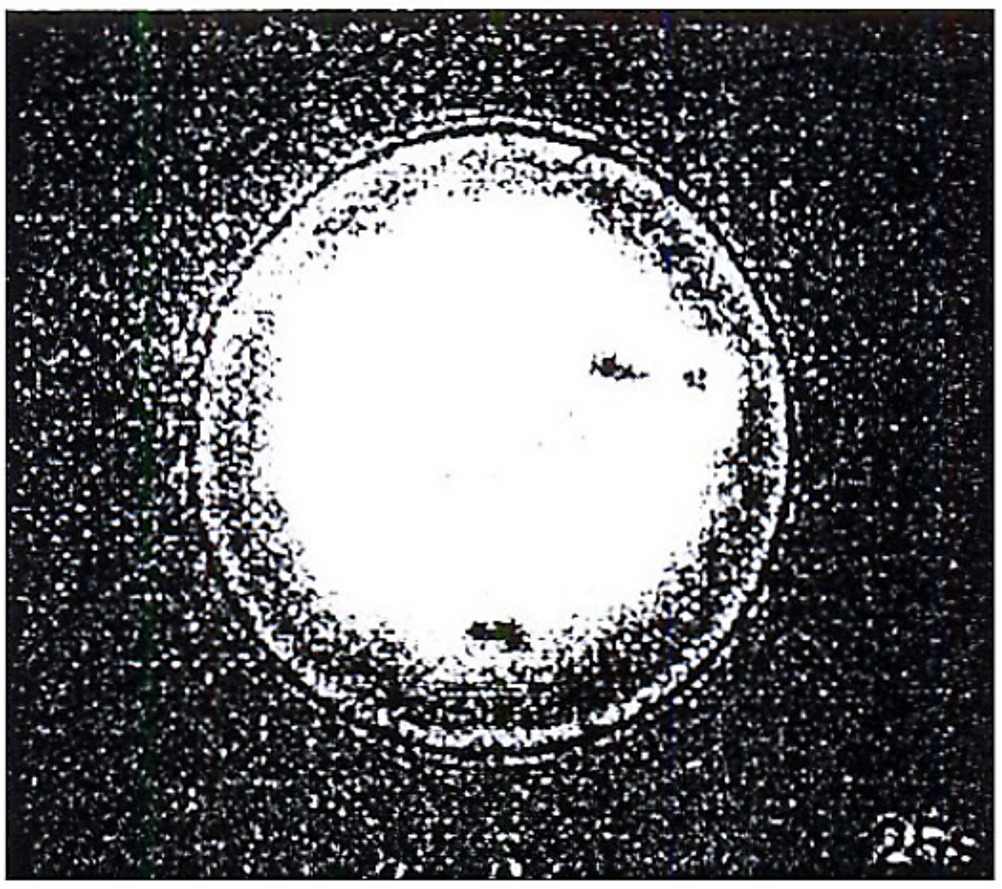
of the molecular charge is provided by the fact that the contact angle varies with pH and passes through a maximum around the isoelectric point, p*I*.

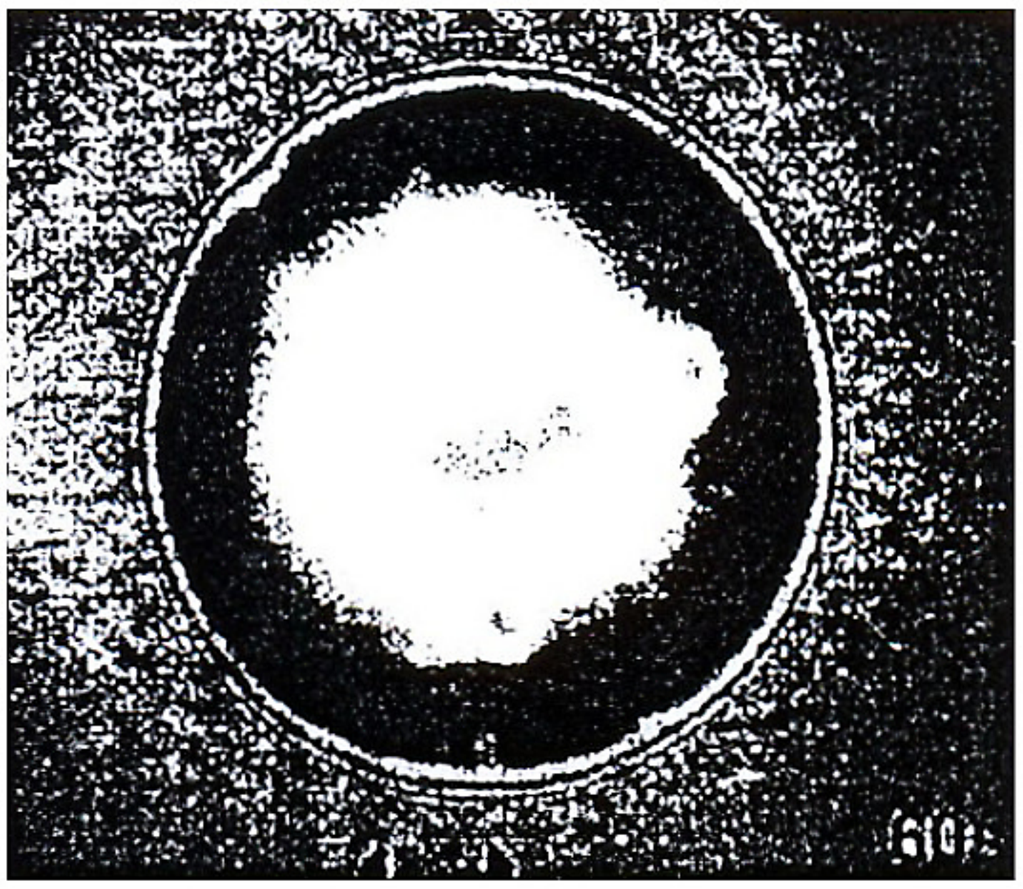
Electrostatic interactions are also observed in thin films stabilized by BLG and β-casein. BLG behaves similarly to BSA although the BLG interfacial behaviour appears to be more pH sensitive near its p*I* (p*I* = 5.4). The BLG emulsion films containing 0.15 M NaCl at pH $\approx$ 6.2 were found to be highly unstable, with rupture occurring almost immediately after the appearance of black spots. Thin films stabilized by β-casein are also sensitive to ionic strength (thick films are obtained without added salt, while the presence of 0.15 M NaCl allows Newton black films to form.[20,21] This electrostatically dominated behaviour is qualitatively similar to prior observations by other investigators, and it effectively validates the Scheludko cell approach for the use of protein-stabilized thin films as models for droplet–droplet interactions in emulsions.

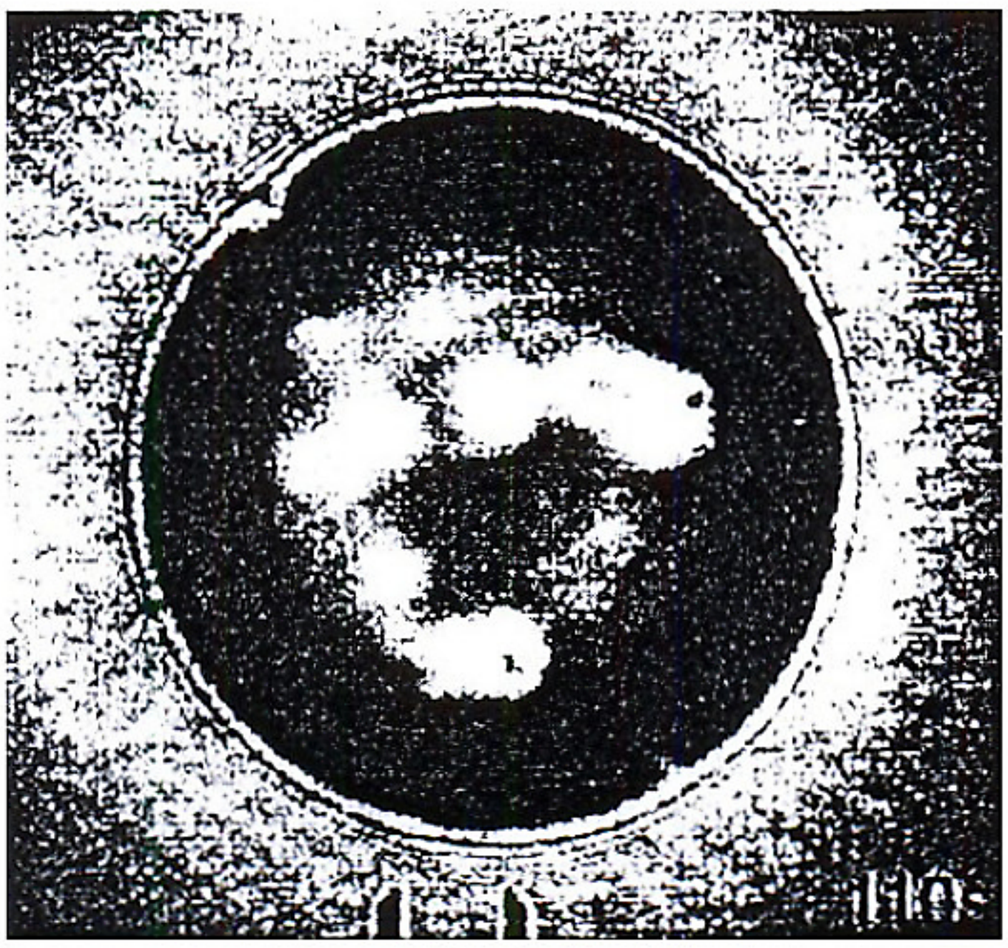
New findings obtained from the above mentioned technique include the establishment of the involvement of protein aggregates in the thinning process. The precise behaviour of these aggregates during film thinning is dependent on protein type and solution conditions. For example, BLG aggregates at pH $\approx$ 6.2 seem to be essentially stiff and they cannot be broken down under the action of the capillary pressure inside the film. However, the gradual disintegration of large protein clusters and an increase in the contact angle accompanied the formation of highly stable BLG Newton black films at pH $\approx$ 7.0. The process is illustrated in Figure 5, where a sequence of photographs shows the time course of protein aggregate disappearance. Finally, after nearly five minutes, the whole film thins to a Newton black film. In the case of β-casein, it is well known that aggregation occurs in solution at room temperature. Any protein lumps caught in black emulsion films were observed to be readily disintegrated by the capillary pressure, which indicates that the aggregation is reversible and that the interfaces are mobile.

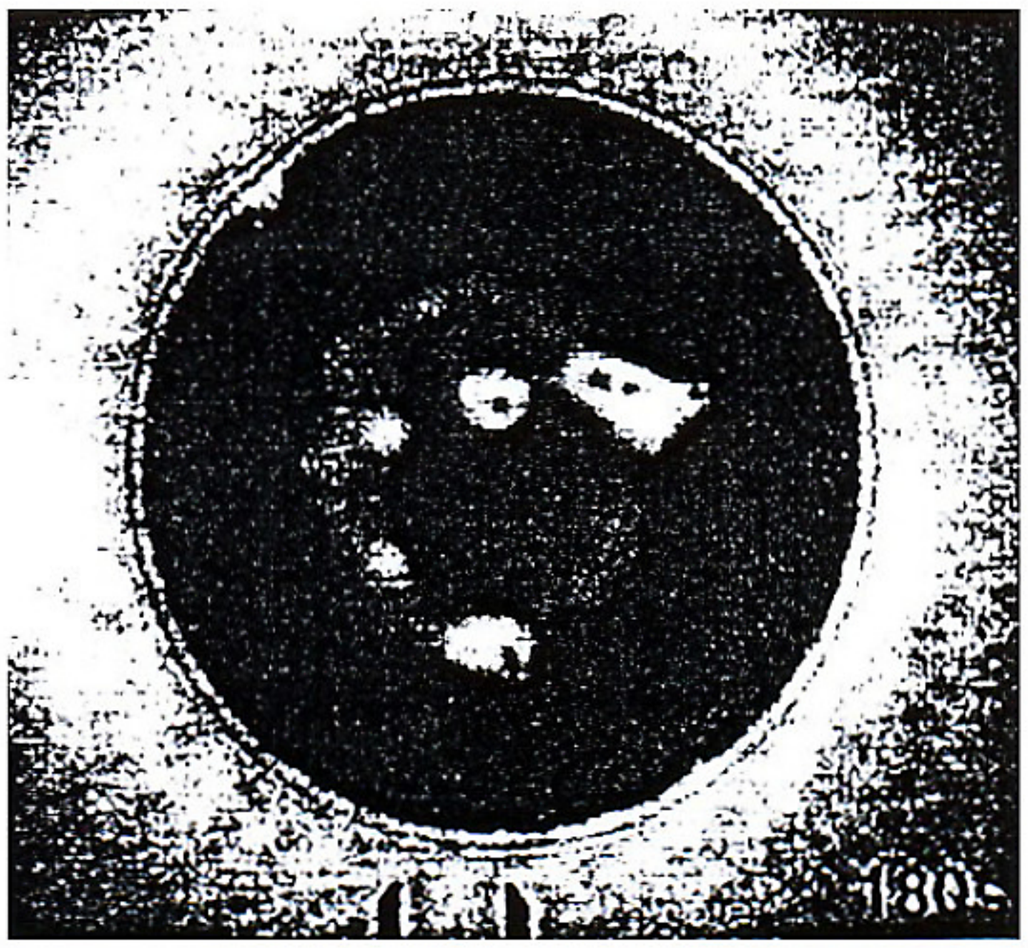
Of the proteins investigated, β-casein has some unique properties which can impact on interfacial behaviour, *e.g.*, its high affinity for Ca^{2+} ions. When the film thickness is measured interferometrically,[20,21] a decrease from $\sim$20 to $\sim$10 nm is observed when the Ca^{2+} concentration is raised above 10 mM (Figure 6). The effect may be attributed to compression of the protein adsorption layers (which has been confirmed to happen at a single interface),[20] or to partial desorption. At the same time, the energy of adhesion between the two interfaces of a Newton black film increases enormously.[21] The cross-binding (at 0.01 wt% casein and 20 mM $CaCl_2$) is so strong that the film surface cannot be detached on applying the maximum capillary pressure accessible in the conventional capillary cell.[21]

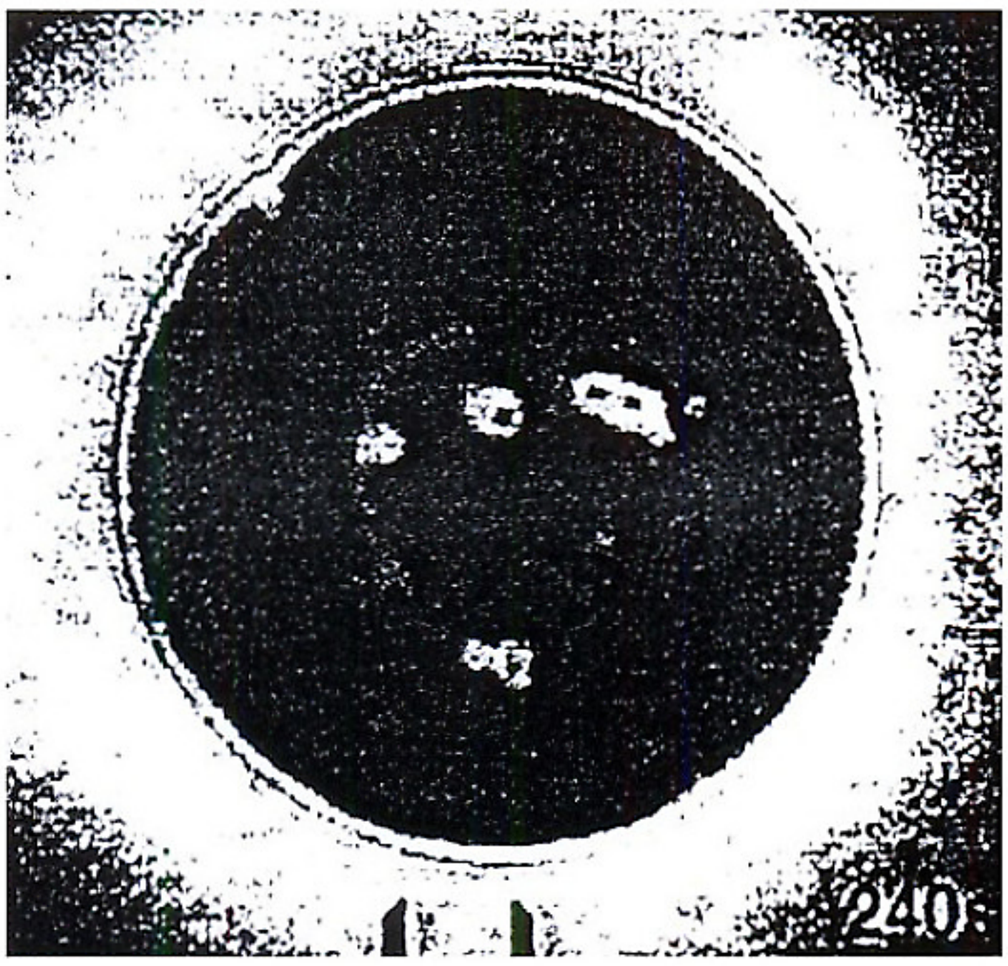
Figure 5 *Interferometric images taken at six consecutive moments during thinning of an aqueous film of β-lactoglobulin in the presence of 0.15 M NaCl and 0.001 M phosphate buffer at pH 6.9. Progressive destruction of protein lumps is observed and finally a Newton black film forms. The distance between marks corresponds to 30.25 μm.*











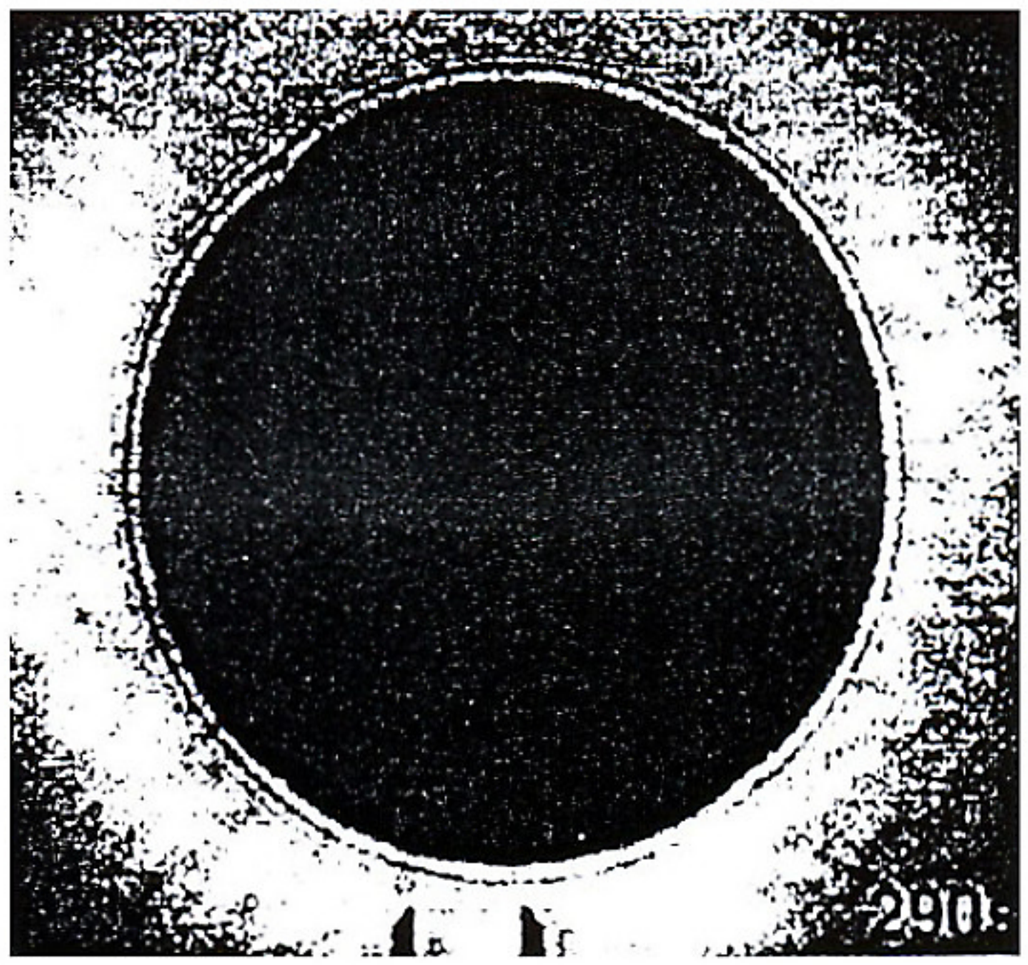


Figure 5

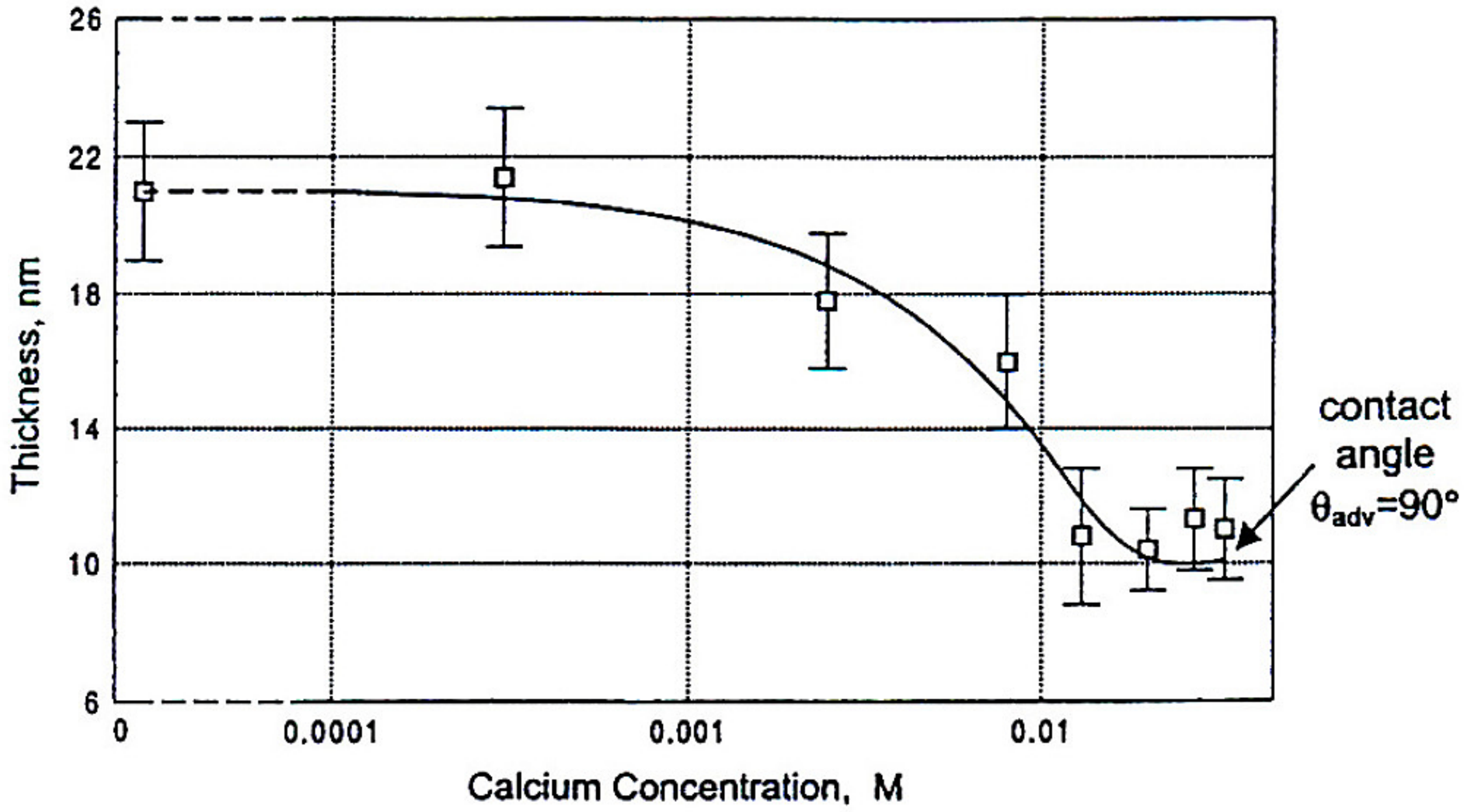


Figure 6 *Thickness of emulsion films with 0.01% (wt) β-casein in the presence of Ca^{+2} ions. Ionic strength* I = 0.15 *M (adjusted by NaCl), pH = 5.0.*

Utilizing the thin film approach, interfacial ageing effects of adsorbed proteins have been characterized by Marinova *et al.*[20] Thin film ageing is investigated by comparing fresh thin film formation (studied immediately after introducing and exposing the oil surfaces to the protein solution) with aged film formation (in which the film is formed 30 minutes after exposing the oil surface to the protein solution). Thin film systems stabilized by BSA in the presence of 0.15 M NaCl were observed to exhibit pronounced ageing effects. Freshly made films typically form very thin, plane-parallel Newton black films. Films formed after 30 minutes ageing contain protein aggregates of irregular shape. Unlike BLG and *β*-casein, which, under the conditions described above, tend to aggregate both in the bulk and on the film surface, BSA aggregates do not occur in the bulk as confirmed by dynamic light scattering. Therefore, the presence of these aggregates are due to aggregation *at the surfaces*. This aggregation is seen to be reversible. Initially the films formed from aged systems thin down and entrap liquid and protein lumps. Subsequently, excess material is gradually squeezed out, and the particles are dispersed under the action of the capillary pressure.

A theoretical explanation of this phenomenon has recently been proposed.[22] This is consistent with the action of surface diffusion as the underlying mechanism for protein disaggregation, as shown by the good fit obtained between theory and experiment. From the best fit one finds the maximum adsorption along the rim where the lump is attached to the interface. The results suggest the existence of protein multilayers at the liquid surface.

Another manifestation of the ageing effect is the hysteresis of the contact angle and its time dependence. The time dependence of the contact angle θ was measured for BSA-stabilized films for more than one hour after loading the two phases in the cell.[20] At fixed capillary pressures, the value of θ was found to keep

on increasing for approximately one hour in an open film (for example, from $\theta = 0.57°$ up to $\theta = 0.8°$ at pH = 6.4). After that, a more or less constant angle is reached. In addition, well pronounced hysteresis of θ develops in these films upon film separation. Hysteresis in these systems is observed experimentally by pushing liquid into the meniscus, which in turn reduces the capillary pressure until the contact line starts shrinking. In this manner one determines the advancing angle, θ_{adv}, which characterizes the force required to detach the adhered film surfaces. The initial value of θ_{adv} with 0.015 wt% BSA is estimated[20] to be *ca.* 2°. After ~30 minutes, a substantial increase of θ_{adv} is such that its measurement becomes impossible, since the Newton fringes around the film periphery cannot be discerned. These films are 'solid-like', *i.e.*, the two interfaces are stuck firmly and irreversibly. In rheological terms, one would say that a 'yield stress' should be applied in order to disjoin the surfaces. The corresponding energy of adhesion thus increases sharply with ageing. Some numerical calculations for the interfacial shape in the transition zone film/meniscus are underway, which will give use the opportunity to evaluate the line tension in films stabilized by proteins. Let us mention here that the notion of 'equilibrium angle' is meaningful only as long as the surfaces retain their fluid nature. Only then can the film diameter adjust for the respective values of the contact angles and capillary pressures in the cell.

3 Structure Formation and Stability of Food Emulsions and Foams

The research discussed above is based on studying two isolated droplets or bubbles and the intervening film between them. It is anticipated that the results from such systems can be applied to understand the behaviour of emulsions and foams as a whole. Recently, we have also pursued a macroscopic approach where the structure of the emulsion and/or foam can be probed directly. Kossell diffraction or 'back' light scattering has been known for many decades. Earlier information obtained by the technique was qualitative. We have recently developed an experimental technique based on this phenomenon that allows the obtaining of quantitative information about the structure of food systems in a non-invasive way without elaborate need for sample preparation. This technique involves shining a monochromatic laser beam on a sample in a glass cell and measuring the back light scattering using a vertically polarized CCD digital camera. Digital signal processing allows for the diffraction pattern to be analyzed to give the particle dispersion structure. The technique was developed and tested on model systems[23] before we ventured into food systems of interest. Again, we studied the frozen whipped topping system.

We have measured the effects of sodium caseinate on fat particle packing structure.[24] The results of back light scattering experiments on a 5.4% fat emulsion system containing two different levels of sodium caseinate are shown in Figure 7. The higher peak of the sample containing twice as high a sodium caseinate level as the other sample indicates that increasing sodium caseinate

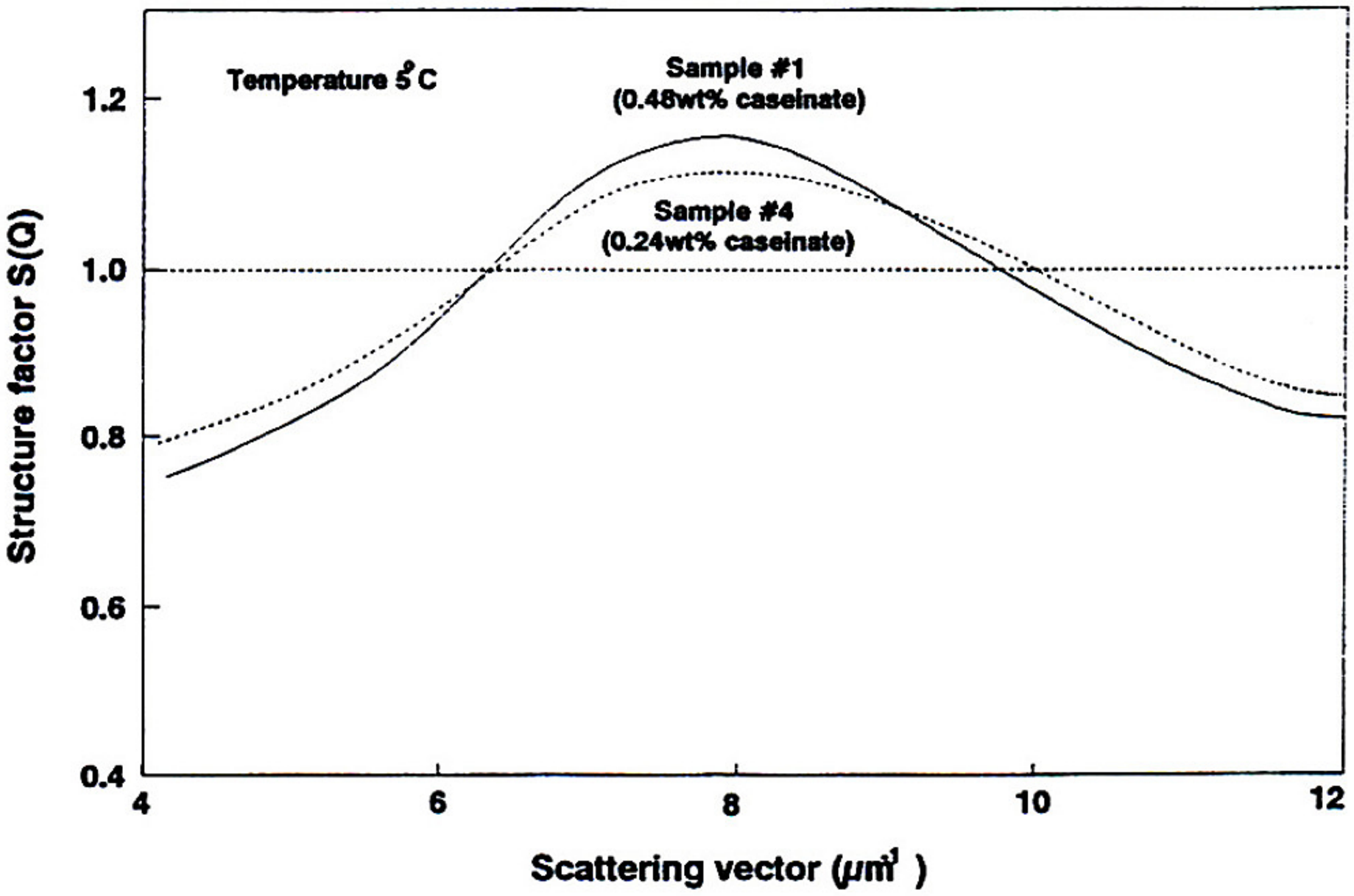


Figure 7 *The effect of caseinate on the fat particle structure inside food emulsions (fat concentration = 5.14%).*

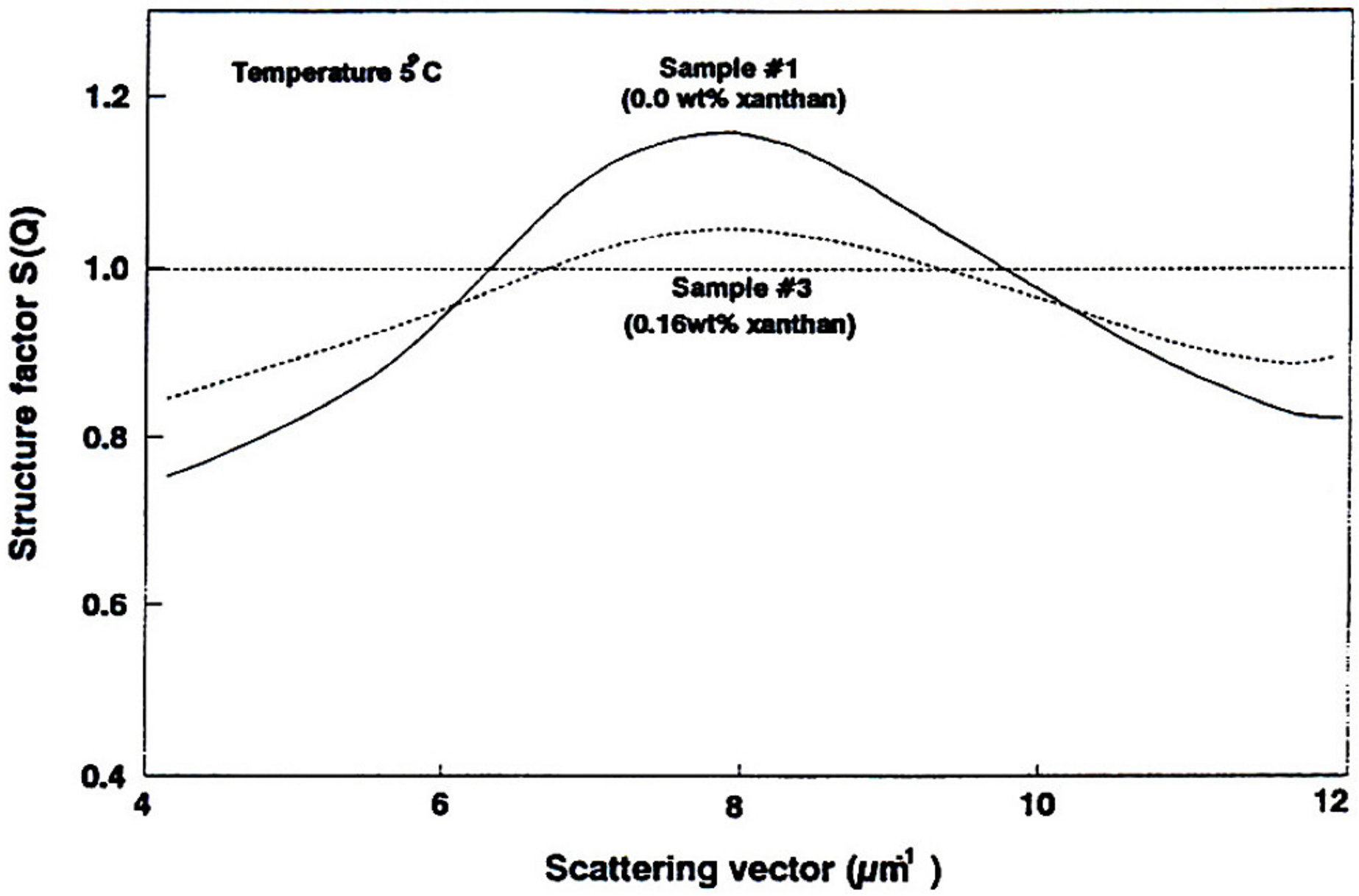


Figure 8 *The effect of xanthan on the fat particle structure inside food emulsions (fat concentration = 5.14%).*

concentration facilitates fat particle structure formation. This is consistent with the observed stabilization of emulsions by sodium caseinate sub-micelles.[17] Theoretical calculations show[24] that the structural energy barrier can be much larger than $3\,kT$ when sodium caseinate sub-micelle concentration reaches 20%.

The effect of xanthan gum on food emulsion stability has also been studied. As shown in Figure 8, the peak height of the sample without xanthan was much higher than that of the sample with xanthan indicating destabilization of the emulsion by xanthan gum. Based on the observation that flocculation of emulsions by xanthan gum can occur at concentrations as low as 10–20 ppm, Koczo *et al.* have argued[25] that the destabilization by xanthan molecules here is by a different mechanism than the depletion flocculation mechanism suggested by Cao *et al.*[26] Rather, they showed the destabilization to be similar to the phase separation of geometrically incompatible polymers demonstrated by Flory.[27] Here, the xanthan molecules (anisotropic phase of elongated xanthan molecules with high aspect ratio) are incompatible with the emulsion droplets (isotropic phase).

The back light scattering method has also been applied to demonstrate fat particle structuring occurring during the whipping process when such an emulsion is whipped in a two-stage process. As measured by the normalized structure factor value, the fat particle structure was improved after each successive whipping stage. A well developed fat particle structure between the air cells is crucial to the stability of the whipped topping.

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