Neutrons, X-Rays and Light

Scattering Methods Applied to Soft Condensed Matter

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(CPCl). Strong electrostatic interactions arise: in spite of the dilution the peak is sharp and the scattering at the beam stop is low. In Fig. 11 the concentration is twice as large but no CPCl is added: there is no electrostatic interaction between the neutral bilayers. Only weak steric interactions remain. The peak is broad and soft and the intensity scattered at beam stop almost diverges. More quantitative procedures have been worked out along these line in order to derive quantitatively the interlayer interactions (Nallet et al., 1993; Zhang et al., 1996; Castro-Roman, 1999).

6.2. L4: an equilibrium dispersion of polydisperse vesicles

The scattering patterns of samples of the L4 phase show nothing specific and provide no direct evidence for vesicles. An example is shown in Fig. 12. Of course, at high q's we find the leading q^{-2} dependence characteristic for the locally bilayered morphology. But at lower q's, there is a progressive crossover towards a weaker dependence which depends in a non-straightforward manner on the system and the concentration of the sample. Therefore, the first evidence of vesicles in the L4 phase was obtained by the Bordeaux group (Herve et al., 1993) from a clever combination of viscosimetry, conductimetry and quasi-elastic light scattering and using 'direct visualization' by freeze fracture electron microscopy. The main reason for the non-specific character of the static scattering is the size polydispersity. Neglecting interactions, for a monodisperse dispersion of vesicles of internal radius R_i and external radius R_e (so $R_e - R_i = \delta$), we would expect for the scattered intensity:

$$I(q) = \phi V(\Delta\rho)^2 \left(3\frac{\sin qR_e - qR_e \cos qR_e - \sin qR_i + qR_i \cos qR_i}{q^3(R_e^3 - R_i^3)} \right)^2 \tag{27}$$

Eq. (27) indeed shows a series of maxima and minima when $q^2 I(q)$ is plotted versus q, characteristic of the two radii: R_e and R_i. But for the very polydisperse situation typical for the L4 phase, these maxima and minima are blurred out and nothing specific remains to be exploited in the scattering. Static scattering alone cannot characterize unambiguously phases of polydisperse vesicles.

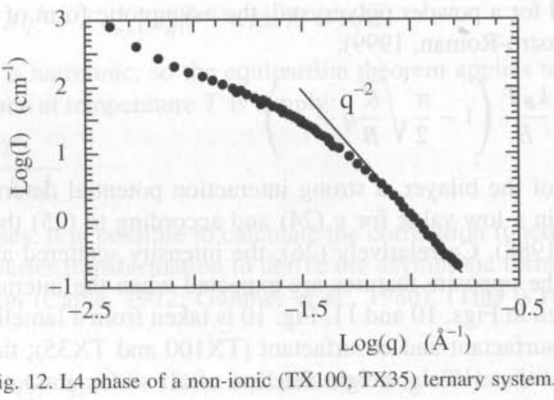

Fig. 12. L4 phase of a non-ionic (TX100, TX35) ternary system.

3.1. Form factors

In the following it will be assumed that the particles are randomly oriented in the sample so that the theoretical form factors for anisotropic particles have to be averaged over orientation. Note that for spherically symmetric objects the form factor can be written as $P(q) = F^2(q)$, where $F(q)$ is the amplitude of the form factor.

1. Homogeneous sphere: The form factor amplitude of a homogeneous sphere was calculated already in 1911 by Lord Rayleigh. For a sphere with radius R:

$$F_1(q,R) = \frac{3[\sin(qR) - qR\cos(qR)]}{(qR)^3}. \tag{21}$$

Sheu (1992) has also given analytical expressions for polydisperse spheres with the number distribution given by, respectively, a box function, a triangular function, an exponential distribution, a Gaussian distribution, a log-normal distribution, and a Schulz–Zimm distribution.

Bagger-Jörgensen et al. (1997) and Svaneborg and Pedersen (2002) [1] have given expressions for the form factor of a sphere with a scattering length density, which gradually decrease at the sphere surface following a half-Gaussian distribution (see form factor 39 and the example in Section 4 for an approximate expression).

2. Spherical shell: This form factor amplitude is obtained by subtracting the empty core with a proper weighting by the volumes:

$$F_2(q) = \frac{V(R_1)F_1(q,R_1) - V(R_2)F_1(q,R_2)}{V(R_1) - V(R_2)}, \tag{22}$$

where $V(R) = 4\pi R^3/3$ and R_1 and R_2 are the outer and inner radius of the shell, respectively. An infinitely thin shell with radius R has the form factor $F_2(q) = \sin(qR)/(qR)$.

Gradzielski et al. (1995) have given the expressions for a shell with a Gaussian radial scattering length density profile.

3. Spherical concentric shells: This form factor amplitude is a generalization of the shell form factor. Let R_i, $i = 1, \ldots, N$ be the radii of the shells and ρ_i be their scattering densities. With this:

$$F_3(q) = \frac{1}{M_3}\left[\rho_1 V(R_1)F_1(q,R_1) + \sum_{i=2}^{N}(\rho_i - \rho_{i-1})V(R_i)F_1(q,R_i)\right], \tag{23}$$

where M_3 is the scattering mass or scattering volume of the particle, given by:

$$M_3 = \rho_1 V(R_1) + \sum_{i=2}^{N} V(R_i)(\rho_i - \rho_{i-1}). \tag{24}$$

Förster and Burger (1998) have recently considered core-shell particles with an algebraic radial scattering length density profile which is piecewise of the form $\rho(r) \propto r^\alpha$. The form factors are given in terms of hypergeometric functions.

[1] Some errors in the paper by Bagger-Jörgensen et al. (1997) are corrected.