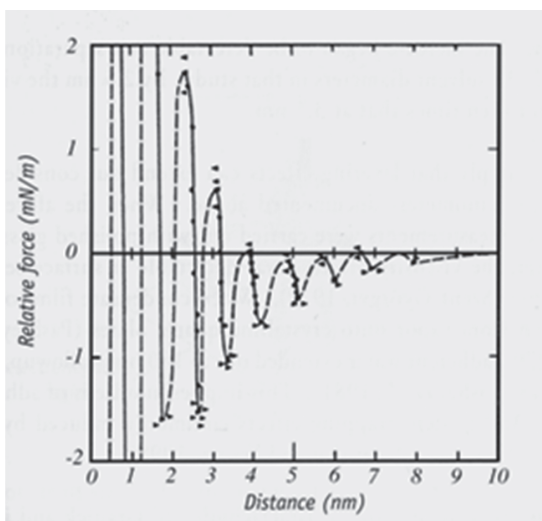


network while still retaining the essential connectivity within the network. Transient hydrogen bonded clusters are permanently formed and broken and very few free water molecules are present, as confirmed by IR experiments (Maréchal 1996). In most simulations, water appears as a three-dimensional network of randomly distributed hydrogen bonds with a local tetrahedral geometry. One of the main questions still open regarding water structure is the way in which hydrogen bonds fluctuate to enable the structural changes to occur. It then seems, as stated by Dore (Dore 2000), that “our fundamental understanding of the disordered hydrogen-bonded network is still at a fairly rudimentary stage”.

The presence of a solid interface in water, here represented by the polymeric network, clearly represents a major perturbation to the hydrogen-bond network. There are several methods used to determine the characteristics of interfacial water (Matricardi et al. 2016):

- a) Measurements of attractive or repulsive forces between two surfaces distant one from another. On this respect Israelachvili and colleagues (Israelachvili and Adams 1978) measured the force required to displace solvents sandwiched between parallel mica surfaces. The closer the surfaces got, the higher the force required. The force-separation relation was not purely monotonic, but a series of regularly spaced peaks and valleys of force were observed. The surfaces have the capacity to organize solvent into at least 10–12 layers (Fig. 5).
- b) Contact angle method
- c) Differential scanning calorimetry
- d) Thermally stimulated depolarization current (TSDC) (Gun'ko et al. 2005)
- e) Dielectric relaxation spectroscopy
- f) Temperature-programmed desorption (TPD)
- g) FTIR and Raman spectroscopies



**Fig. 5.** Organization of the solvent in 10–12 layers as described by the force separation relation (Israelachvili 1978).