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The dynamics of hydrophobic interaction

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SUMMARY

To investigate the relation between the structure of liquid water and hydrophobic interaction the dielectric relaxation of water in dilute aqueous solutions has been studied. After an introduction of the problem in which the dynamic aspects of water structure are emphasized, a theoretical analysis of the dielectric relaxation of water and aqueous solutions is given (Chapter 1.3). It can be shown that dielectric difference measurements at microwave frequencies are very useful to study changes in water structure resulting from the dissolution of (hydrophobic) solutes.

The transmission of electromagnetic radiation through dielectrics with high loss and through waveguides is discussed on the basis of Maxwell theory in Chapter 2. Section 2.3 presents a summary of the theory of cavity resonance perturbation, which is the basis of our experimental methods. Several important aspects of the microwave system, which is able to discriminate the dielectric difference between water and a 1 mM salt solution, are described in Chapter 3. A careful analysis of experimental results in terms of cavity perturbation theory is given in Chapter 4.

The assumption that dielectric relaxation parameters depend linearly on solute concentration has been studied carefully in Chapter 5.1.

A study of Helix Pomatia hemocyanin solutions at different dissociation stages of the protein (Chapter 5.2) did not give more information than a hydration number, which appeared to be somewhat larger for tenths and twentieths than for halves and undissociated molecules.

In addition experiments on a series of largely hydrophobic solutes like higher alcohols and carboxylic acids (up to hexanols and hexanoic acid) in dilute aqueous solution have been carried out. The dielectric measurements (Chapter 6.2.1 and 6.3.1) are interpreted as a shift in water relaxation frequency and a decrease of the static dielectric constant. An alternative interpretation in terms of a mixture containing water with an unchanged relaxation frequency and a solute with an additional relaxation could be ruled out (Chapter 7.1).

Further conclusions from the dielectric measurements were the absence of tightly bound water and the fact that the spread in water relaxation frequencies was not larger in the solutions than in pure water. On this basis an exchange process between water in the first hydration layer around hydrophobic solutes and water in the bulk is proposed, which is faster than the dielectric relaxation rate (Chapter 7.2; see also Chapter 5.1.3).

A correlation between the large negative standard entropies of transfer for hydrophobic solutes and the changes in dielectric relaxation frequency has been found (Chapter 6.4). A model of water relaxation on the basis of a hindered rotator (Chapter 7.3) shows that about 15 percent of the experimental transfer entropies are sufficient to bring about the relaxation frequency shifts actually observed. The major part of the entropy effect will probably be due to a hindering of translational motions, which are also slowed down. We can almost certainly conclude, however, that the entropy effects underlying hydrophobic interaction are of dynamic nature and can not be ascribed to the existence of highly immobilised cages of water molecules around hydrophobic groups.