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III. MACROMOLECULAR POLARITIES

A. FREE MACROMOLECULES

1a. Deoxyribonucleic acid (DNA)

DNA mobility and dynamics steady-state fluorescence polarization have been studied by Haerd (1986).

In the contact between DNA and binding proteins positively charged amino acids interact with DNA backbone or basis (Harrison and Aggarwal, 1990).

a³ Transcription

Intracistronic polarity is the preferential expression of the promotor-proximal part of a gene as described in *Escherichia coli* (Hansen *et al.*, 1973). The reduced expression of promotor-distal parts of an operon relative to the promotor-proximal parts has been observed in many situations where translation is hindered. Exposition of *E. coli* cells to a nutritional downshift leads to the cessation of stable RNA accumulation. Following such downshift, a strong polarity was observed for the transcription of *lacZ* (Johnsen *et al.*, 1977) indicating that RNA polymerase runs away from the leading ribosomes in ppGpp synthetase mutants (Jensen and Pedersen, 1990).

2. PROTEINS

Helices in proteins are typically involved in multiple hydrogen-bonded, van der Waals, and electrostatic interactions. Constitutive amino acids have distinct conformational differences that lead to stabilization or destabilization of an α -helix (Fasman, 1989).

The α -helix dipole finds its origin in the dipoles of the single peptide units. Its formal charges lead to a dipole moment. Hydrogen bonds cause this polarization and thereby the peptide dipole moment of the α -helix. The field of a continuous line dipole is equal to the field of a positive charge at the amino end and a negative charge at the carboxyl end. The helix dipole interacts with charged phosphate at N-termini and thus might be used in catalysis (Hol *et al.*, 1978).

Important for such helix stabilization are electrostatic interactions between charged side-chains and either another charged residue or the helical dipole (O'Neil and DeGrado, 1990). It is the arrangement of positive and negative charges along the helix which affect helix stability by an interaction between charges and the helix dipoles (Wada, 1976; Hol, 1985).

There are many models of protein folding (see Wetlaufer, 1990) possibly involving

structural constructs such as nucleation sites and flickering clusters, a term adopted in the field of water structure. Protein folding patterns require a compact structure and hydrogen bonds formed by buried polar groups. They necessitate the formation of α -helices or β -sheets which assemble to give the molecules their globular three-dimensional structures. The usual antiparallel packing (180°), rather than parallel manner (360°), of pieces of secondary structures that are adjacent in the protein sequence is one feature of chain topology (Chothia and Finkelstein, 1990).

Polarity (or lack of it) determines the nature and strength of interactions between amino acids in a protein and between the protein and water. The differences among amino acids stem from differences in their side chains namely, in shape, size and polarity. Shape and size affect the packing together of amino acids in the final molecule (Richards, 1991).

3'. POLYENES

These hydrocarbons are highly polarizable; that is electric field can induce substantial dipole moments (see also IV.D.3). Direct determinations of electro-optic parameters for these polyene chromophores by Stark effect spectroscopy have provided some quantitative basis in the evaluation of carotenoid band shifts under physiological conditions (Gottfried *et al.*, 1991). The centrosymmetric carotenoids in organic solvents show apparent dipole moment difference along with large changes in polarizability. Symmetry-breaking perturbations in solvents might be the reason for the apparent excited state dipole moment of these polyenic compounds (Liptay *et al.*, 1988).

5. ENZYMES

Protein structure and enzyme function are the finely balanced end-products of weak interactions. Proteins are built on definable principles and enzymes use recognizable catalytic devices. In an extensive review, Hol (1985) proposed that the formal dipole associated with the classical α -helix modulates the properties of groups at the helix termini. The effects might be due to the electric field of α -helix dipole. From mechanistic and structural considerations on triosephosphate isomerase which imply polarity interactions, Knowles (1991) speculates that "the relatively large size of enzymes compared with most of their substrates may derive from the need for a matrix that positions their functional groups, focuses their helices and anchors the ends of their mobile loops".

In the process of lipase-catalyzed transesterification and esterification, polarity of every substrate must be obligatorily considered because of its ability to modify the water partition between the solid phase (enzyme) and the liquid phase (substrate and product) thereby leading to drastic changes in enzyme activity (Goldberg *et al.*, 1990).

6. ANTIGENS-ANTIBODIES

Antibody-antigen interaction involves molecular recognition and its specificity is not only made by shape complementarity such as depressions on one surface field by protuberances from the other but by hydrogen bonding. Of particular relevance here is the directionality of the hydrogen bonds, necessitating a hydrogen bond receptor within a certain distance and within a certain solid angle of the hydrogen bond donor in order to form a strong bond. Antibody-antigen interfaces differ, with an average of about a dozen hydrogen bonds. The protein surface that is combining with the antibody has a significant content of polar residues (Davies *et al.*, 1990).

7. SYNTHETIC POLYMERS

Mixtures of deuterated and normal - protonated - polymers are chemically indistinguishable. However, above a certain molecular weight, symmetric mixtures of deuterated and protonated polybutadienes phase separate (Bates *et al.*, 1985), a universal phenomenon which is a consequence of the well-known reduction in carbon-hydrogen bond length that accompanies deuterium substitution in organic molecules. Such a decrease of the bond length reduces the bond polarizability, which is manifested as decreased segment polarizability (Bates, 1991).

The first step towards electronic devices with a polymer playing an active role was described in 1988 by Bloor and by Garnier *et al.* (1990) who report the construction of an all-organic transistor based on sexithienylene. Recombination of electrons and holes at the interface between emitter and transport layers produces an electroluminescence of high intensity. Among the possible mechanisms for electroluminescence, there are two possibilities, namely that of formation of negative and positive polarons would rather combine to form a neutral polaron exciton or that of electrons which would tunnel directly to the exciton levels (Bloor, 1991).

Characterization of electroluminescence in the conjugated polymer poly(*p*/phenylene vinylene) or PPV is also of interest to understand the fundamental excitations in this class of organic semiconductors. For such polymers, these excitations are polarons, either uncharged, as the polaron exciton, or charged, singly charged as the polaron, and doubly charged as the bipolaron (Fesser *et al.*, 1983). For electroluminescence, bipolarons, paired charges with low mobility, are very unlikely to be the charge carriers responsible for formation of polaron excitons. Therefore the charge carriers involved in the process are probably polarons (Burroughes *et al.*, 1990). This idea contrasts with the results from the previous optical work (Friend *et al.*, 1987) which suggest that bipolarons are the most stable quasiparticles responsible for the conductivity in PPV.

B. AGGREGATES

1. Crystals-quasicrystals

Contrarily to metal crystals, quasicrystals such as dodecahedral grains of aluminium, copper and iron melted together and cooled (Nelson, 1986, see I) cannot be

constructed from atoms in repeated unit cells. Laboratory produced quasicrystals conduct electricity rather poorly (Stephens and Goldman, 1991).

2. Viral assemblies

c) *Polar viral morphopoiesis*

DNA replication and regulated transcription in the development of bacteriophage T4 are closely connected (Epstein *et al.*, 1963). The enhancer of T4 late transcription is a break in the nontranscribed DNA strand to which the DNA polymerase accessory proteins bind. The noted preference of these polymerases for the nontranscribed strand point to a “polarity” for the placement of the nicked DNA template and present “an interesting puzzle concerning one aspect of the mechanism of action of the DNA polymerase accessory proteins” (Herendeen *et al.*, 1990). However, because of close coupling between recombination and replication in this phage development, there is no distinguishing relation polarities of late transcription and replication (Mosig, 1987).