rotational diffusion membrane and fluorescence anisotropy

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Structural properties of model membranes, such as lipid vesicles, may be investigated through the addition of fluorescent probes. After incorporation, the fluorescent molecules are excited with linearly polarized light and the fluorescence emission is depolarized due to translational as well as rotational diffusion during the lifetime of the excited state. The monitoring of emitted light is undertaken through the technique of time-resolved fluorescence: the intensity of the emitted light informs on fluorescence decay times, and the decay of the components of the emitted light yield rotational correlation times which inform on the fluidity of the medium. The fluorescent molecule DPH, of uniaxial symmetry, is rather hydrophobic and has collinear transition and emission moments. It has been used frequently as a probe for the monitoring of the fluidity of the lipid bilayer along the phase transition of the chains.
The interpretation of experimental data requires models for localization of fluorescent molecules as well as for possible restrictions on their movement. In this study, we develop calculations for two models for uniaxial diffusion of fluorescent molecules, such as DPH, suggested in several articles in the literature. A zeroth order test model consists of a free randomly rotating dipole in a homogeneous solution, and serves as the basis for the study of the diffusion of models in anisotropic media. In the second model, we consider random rotations of emitting dipoles distributed within cones with their axes perpendicular to the vesicle spherical geometry. In the third model, the dipole rotates in the plane of the of bilayer spherical geometry, within a movement that might occur between the monolayers forming the bilayer.
For each of the models analysed, two methods are used by us in order to analyse the rotational diffusion: (I) solution of the corresponding rotational diffusion equation for a single molecule, taking into account the boundary conditions imposed by the models, for the probability of the fluorescent molecule to be found with a given configuration at time t. Considering the distribution of molecules in the geometry proposed, we obtain the analytical expression for the fluorescence anisotropy, except for the cone geometry, for which the solution is obtained numerically; (II) numerical simulations of a restricted rotational random walk in the two geometries corresponding to the two models. The latter method may be very useful in the cases of low-symmetry geometries or of composed geometries.