Mitigation of stochastic effects in time-resolved serial crystallography data by low-pass spectral analysis

Time binning approaches have proved successful in dealing with the issues of sparsity and partiality in time-resolved serial crystallography and produced molecular movies that offer unprecedented insight into the function of biomolecules in action. Yet the averaging procedure that underlies such approaches normally requires large numbers of frames: this often means binning over large time windows, which implies a loss of timing information.

While the data manifold of the time-evolving system is intrinsically one-dimensional - and its trajectory likely explores only a low-dimensional subspace of the high-dimensional data space, the physical specificities of the diffraction experiment introduce incompleteness and partiality, artificially increasing the apparent dimension of the subspace in which the data points lie. Time-lagged embedding mitigates such issues and allows - at least in favourable situations - to reconstruct the underlying system dynamics by singular value decomposition in the supervector space (Singular Spectrum Analysis). Nonetheless this procedure is impractical in the case of large data sets, and in some circumstances may not be able to provide a low-rank approximation of the system dynamics [1]. Time-lagged embedding can be combined with data filtering in supervector space to ease these issues. This can be done - for instance, by using a set of data-driven basis functions, in an approach called the nonlinear Laplacian spectral analysis (NLSA) [1,2]. Here we present an alternative approach, which we call the low-pass spectral analysis (LPSA), whereby a set of orthonormal trigonometric functions is used as subspace basis [3]. Compared to NLSA, LPSA presents various important computational advantages, in particular concerning the dimension of the parameter space to be explored.

These concepts are exemplified using synthetic models and preliminary serial crystallography results from the membrane protein bacteriorhodopsin in the first ps after photoactivation.

References

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