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MODELING OF SINGLET–SINGLET ANNIHILATION IN MOLECULAR LATTICE

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Singlet–singlet annihilation is a common phenomenon in molecular structures. It is often difficult to provide experimental conditions that could prevent annihilation; therefore, it might often be necessary to account for annihilation while analyzing data.

Simple annihilation model can be described by rate equation Eq. 1:

$$\frac{\mathrm{d}n}{\mathrm{d}t} = -\gamma n^2, \tag{1}$$

here $n(t)$ is the mean number of remaining excitations in the system at time t and γ is the rate constant for annihilation. In this model, the size of molecular aggregate, excitation transfer rate across the aggregate, and initial population of excitations are considered to be very large. In order to include finite transfer rate, γ must be considered as a function of time, which for one-dimensional systems at longer times can be approximated by a power law [1]. More precise statistical model accounts for the discrete number of excitations, but still considers the whole aggregate as a supermolecule, resulting in the system of Pauli Master equations [2].

To account for both finite transfer rate and discrete number of excitations in annihilation model, we have chosen to use Monte Carlo method. The model is based on continuous time random walk. After sufficient number of lattices have been generated, the average kinetics of specific initial number of exitations is calculated (Fig. 1). The average kinetics is later recalculated considering that initial distribution of excitations obeys a Poisson distribution. The kinetics is then approximated using Eq. 2 to evaluate the $\gamma(t)$ dependence:

$$\frac{\mathrm{d}n}{\mathrm{d}t} = -\gamma(t)n^2 - k_{rel}n. \tag{2}$$

Here k_{rel} is the rate constant for relaxation.

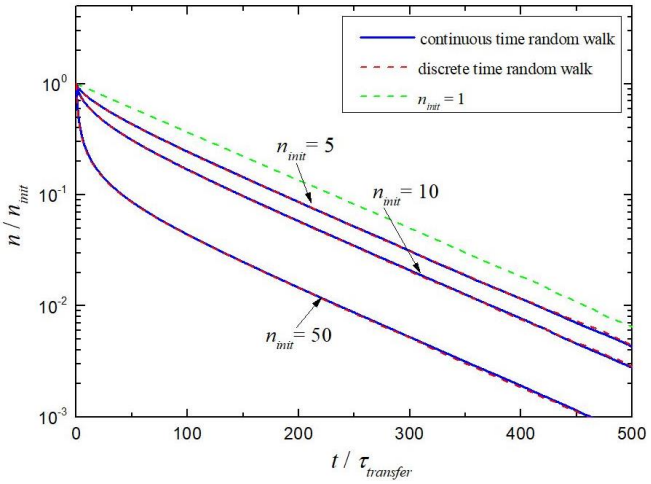

Fig. 1. Excitation population kinetics in one-dimensional aggregate (n_{init} is the initial population size, molecular lattice size is N = 100 nodes). When one excitation is left in the lattice, quenching becomes exponential due to linear relaxation (relaxation rate is $k_{rel} = 10^{-2}\, k_{\mathrm{transfer}}$).

References

1. H. van Amerongen, L. Valkunas, R. van Grondelle. Photosynthetic Excitons. World Scientific, Singapore, 2000.
2. V. Barzda, V. Gulbinas, R. Kananavicius, V. Cervinskas, H. van Amerongen, R. van Grondelle, L Valkunas, Singlet-Singlet Annihilation Kinetics in Aggregates and Trimers of LHCII, Biophys. J. **80** (2001) 2409–2421.