

Non-adiabatic dynamics of light-driven molecular motors for membrane applications

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Recently light-driven molecular nano motors (MNMs) based on overcrowded alkenes (Fig. 2) have been applied to disrupt biological phospholipid bilayers (PLB).[1] This mechanism has potential application in pharmacology combating cancer and infectious diseases, since it can be used to induce cell death or for drug delivery. The temporal and spatial control of light-driven MNMs allows the development of highly selective drugs, minimizing side effects and drug resistance.

However, being still at a developmental stage, a detailed understanding of the molecular mechanism of this process is required to advance this technology towards clinical applications. To reduce phototoxicity, it is necessary that the motor operates with a high quantum yield, converting a high percentage of the absorbed photons into mechanical work to displace membrane lipids. Furthermore, tissue applications require the irradiation wavelength to occur in the 600–1000 nm region, which penetrates deeper than the initially used ultraviolet light, that also has higher phototoxicity.

Here we study the photodynamics of overcrowded alkenes using the time-dependent density functional theory surface hopping (TDDFT-SH).[2] We report wavelength-dependent product quantum yields [3] and excited state lifetimes. Furthermore, we introduce different functional groups to enhance two-photon excitation cross sections to allow for excitation in near infrared and visible region.

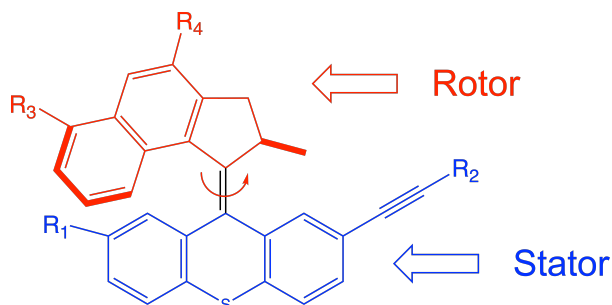


Figure 1: Structure of the MNM. Substituents R_1 and R_2 enhance binding of the MNM to the membrane; R_3 and R_4 can be varied to tune absorption properties.[4].

Index Terms: surface hopping, light-driven molecular nano motors, wavelength-dependent quantum yields

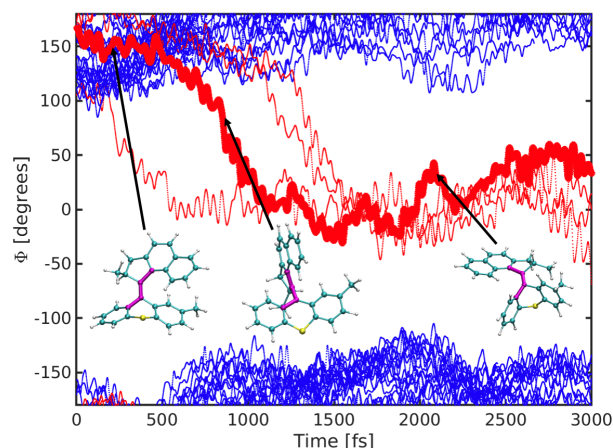


Figure 2: Time evolution of the dihedral angle (magenta), corresponding to the reaction coordinate of the drilling motion, simulated by TDDFT-SH. Out of 50 trajectories, four trajectories (red) lead to a successful 180° rotation. 46 trajectories did not isomerize within the first 3 ps after photoexcitation (blue). Three representative snapshot structures at the indicated simulation times are shown for the trajectory (thick red).

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