First principles prediction of wavelength-dependent isomerization quantum yields of a second-generation molecular nanomotor

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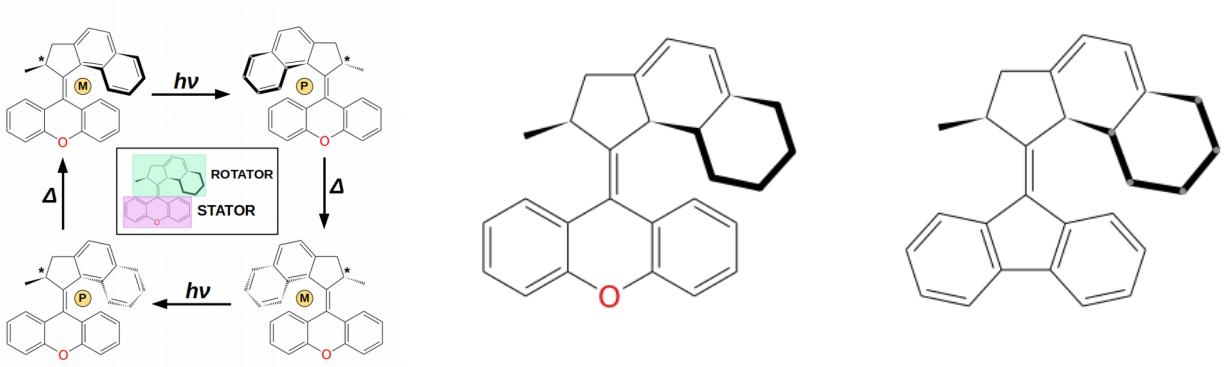
Second-generation molecular nanomotors (MNMs) are gaining interest in biomedical applications due to their potential for light-induced ultrafast photoisomerization [1,2]. The rotational motion of this kind of MNMs is based on a two-step process. First, light absorption induces an E-Z photoisomerization around the central carbon-carbon double bond, converting the molecule into an unstable conformer. This step is followed by a thermal helix inversion, where the molecule undergoes a conformational change that restores the stable conformer but with a different spatial orientation. Repeating these two steps in sequence enables a full 360º unidirectional rotation. A fundamental challenge in their design is achieving unidirectional rotation during isomerization and thermal helix inversion. In this contribution, we aim at determining a way to ensure that unidirectionality [3]. For that, we investigate and compare the excited-state dynamics of all the possible conformers of two different second-generation Feringa-type MNM using non-adiabatic trajectory surface hopping molecular dynamics based on time-dependent density functional theory (TDDFT).

We simulate the photoisomerization of both M- and P-conformers and calculate quantum yields for clockwise and anti-clockwise rotations. Our results reveal that the helicity of the starting conformer determines the isomerization direction. Excitation in the wavelength range where the most stable conformer exhibits maximum absorption and quantum yield, maximizes unidirectional rotation. Additionally, we report excited-state lifetimes and detailed structural dynamics, providing new insights into the photochemical behavior of MNMs. These findings contribute to the rational design of more efficient molecular nanomotors for biomedical applications.



[1] V. García-López, D. Liu, J. M. Tour, *Chem. Rev.*, **2019**, *120*, 79-124.
[2] V. García-López, F. Chen, L. G. Nilewski, G. Duret, A. Aliyan, A. B. Kolomeisky, J. T. Robinson, G. Wang, R. Pal, J. M. Tour, *Nature*, **2017**, *548*, 567-572.
[3] J. Lucia-Tamudo, M. Menkel-Lantz, E. Tapavicza, *in preparation*.