Photoprotecting uracil by coupling with lossy nanocavities

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Abstract

Photodamage in nucleobases is associated to the excitation of the molecule to the S_2 state, typically occurring when the molecule is irradiated by UV light [1]. More practically, the probability to incur photodamage is associated to the long permanence of the nuclear wavepacket on the electronic state S_2 [2]. Strong coupling between molecules and plasmonic nanocavities has emerged in the last few years [3] as a compelling strategy to modify and possibly control photochemical reactions, but is limited by the intrinsically short (few fs) lifetimes of plasmonic nanocavities. In this work, we explore how to make virtue out of necessity by making use of the cavity losses. We perform full quantum dynamics simulations on pre-computed potential energy surfaces to describe the molecular relaxation, accounting for cavity losses by means of a non-Hermitian Hamiltonian. Remarkably, we identify that the optimal photoprotection is achieved when the coupling between molecule and nanoparticle is comparable to the cavity losses, just at the border between the weak and strong coupling regimes.

REFERENCES

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FIGURES

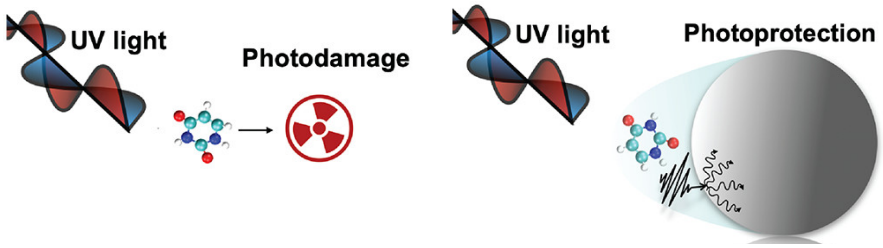

Figure 1: Uracil under action of UV-light may incur in photodamaging reactions, potentially resulting in dangerous mutations. We couple the uracil molecule to a silver nanosphere to open up an efficient relaxation channel, finally achieving a photoprotective effect.

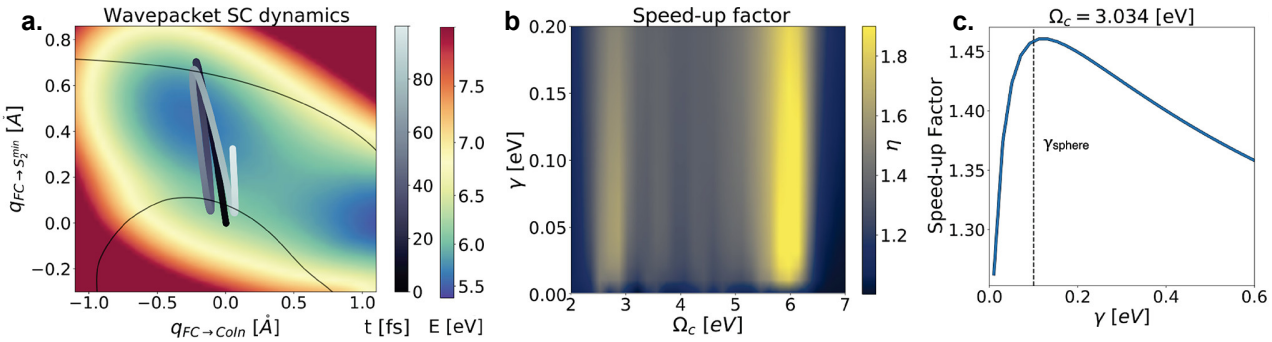

Figure 2: By investigating the nuclear wavepacket dynamics on the uracil potential energy surfaces, we are able to find suitable conditions to stir the wavepacket away from dangerous reaction pathways (Panel **a.**). Quantifying the photo protection effect, we find that the optimal photoprotection (Panels **b.** and **c.**) is achieved at the crossover between the weak and strong coupling regimes.