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(Dated: August 17, 2021)

Theoretical prediction of electronic absorption spectra without input from experiment is no easy feat as it requires addressing all the factors that affect line shapes. In practice, however, the methodologies are limited to treat these ingredients only to a certain extent. Here we present a multiscale protocol that addresses the temperature, solvent and nuclear quantum effects, anharmonicity and reconstruction of the final spectra from the individual transitions. First, QM/MM molecular dynamics is conducted to obtain trajectories of solute-solvent configurations, from which the corresponding quantum corrected ensembles are generated through the Generalized Smoothed Trajectory Analysis (GSTA). The optical spectra of the ensembles are then produced by calculating vertical transitions using TDDFT with implicit solvation. To obtain the final spectral shapes, the stick spectra from TDDFT are convoluted with Gaussian kernels where the half-widths are determined by a statistically motivated strategy. We have tested our method by calculating the UV-vis spectra of a recently discovered acridine photocatalyst in two redox states. Vibronic progressions and broadenings due to the finite lifetime of the excited states are not included into the methodology yet. Nuclear quantization affects the relative peak intensities and widths, which is necessary to reproduce the experimental spectrum. We have also found that using only the optimized geometry of each molecule works surprisingly well if a proper empirical broadening factor is applied. This is explained by the rigidity of the conjugated chromophore moieties of the selected molecules which are mainly responsible for the excitations in the spectra. In contrast, we have also shown that other parts of the molecules are flexible enough to feature anharmonicities that impair the use of other techniques such as Wigner sampling.

I. INTRODUCTION

Photocatalysts employed under homogeneous catalytic conditions are molecules that harvest the energy of visible light to facilitate new transformations and new synthetic routes which may yield otherwise inaccessible scaffolds and molecules.¹ The typical photocatalytic scenario is that upon interaction with light the photocatalyst molecules reach accessible excited states and then they quench rapidly to the lowest available excited state within the same spin manifold (e.g. a singlet molecule will be in the S_1 state). Depending on the circumstances, the further interconversions define the subsequent feasible catalytic processes such as reaching a T_1 (first triplet) state and then engaging in various electron transfers to trigger a photoredox transformation. The molecules typically employed in photocatalysis absorb visible light, a feature which has a significant advantage: they can be selectively excited in the visible region whereas typical organic substrates and solvents absorb in the UV region.

This selectivity can be achieved by inserting and tuning chromophore groups, such as delocalized π -systems. The electronic properties of photocatalysts can be explored by measuring or calculating their electronic (UV-vis) spectra. In this regard, calculations can be very useful because they provide a large amount of information not directly available from experiment; such as the assignment of bands to transitions between electronic states or the identification of dark states. Calculations can also help to understand how structural and electronic modifications introduced to photocatalysts affect the excitations, therefore facilitating the design of new photocatalysts.

These computations, however, have to address a number of challenges. To begin with, the description of the electronic structure in both ground and excited states requires sufficiently accurate methods.² In practice, TDDFT employing functionals with exact exchange contribution is usually adequate,³ but more accurate, wavefunction-based methods should be considered for higher accuracies. Another issue is to capture effects arising from fluctuations induced by the environment such as temperature and solvent. In fact, routine calculations can only provide a crude approximation to these as they use a single configuration to obtain the excitation spectrum.^{4–6} For benchmarking purposes, however, this

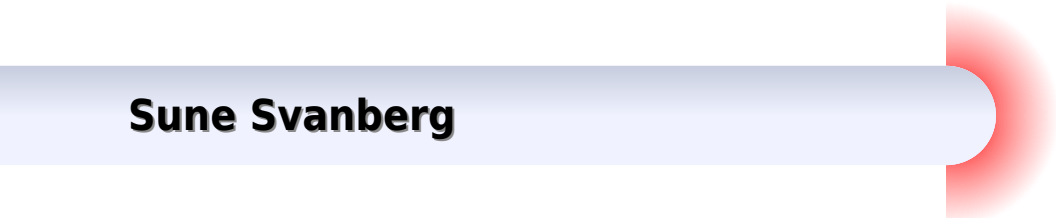
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