

VISTA Seminar

Seminar 8

9:30 – 11:00 am EST / 2:30 – 4:00 pm GMT / 3:30 pm – 5:00 pm Paris

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New Analysis Tools for Excited-State Quantum Chemistry: Turning Numbers into Chemical Insight

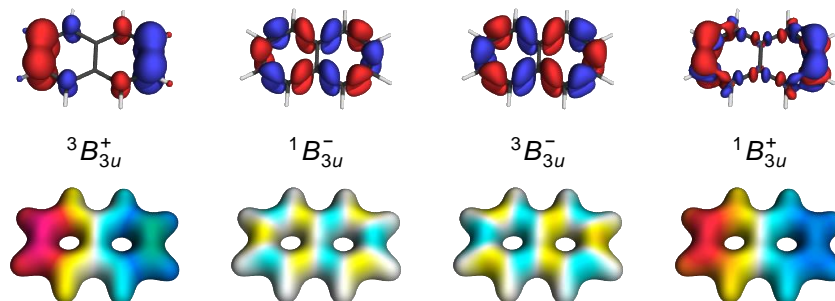
Felix Plasser

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Excited electronic states are central to many areas of modern science and computational methods are becoming ever more accurate in their description. However, a new bottleneck is encountered in the analysis of the computations owing not only to the quantity of data produced but also because many of the phenomena studied are difficult to grasp within the standard molecular orbital (MO) picture at all. To remove this barrier an extended wavefunction analysis framework has been developed over the last years. We will illustrate how these tools can be used to (i) analyze excited state character in an automated and reproducible way [1], (ii) bridge between the MO, exciton and valence-bond pictures, and (iii) provide new insight into excited state energetics beyond the MO picture [2].



As a first example, we illustrate how excited-state localization and charge transfer in transition metal complexes can be quantified in an automated way not requiring any visual inspection of orbitals [3] and show the application of this method in different use cases [4]. Subsequently, we outline how the developed methods provide new insight into the connections of the MO and valence-bond pictures using the paradigmatic case of the ionic and covalent states of naphthalene [5] and present a model for explaining the relative energies of the involved states based on their transition densities [2], see Fig. 1.



TOC: Transition densities and associated electrostatic potentials of the lowest singlet and triplet B_{3u} states of naphthalene.

References

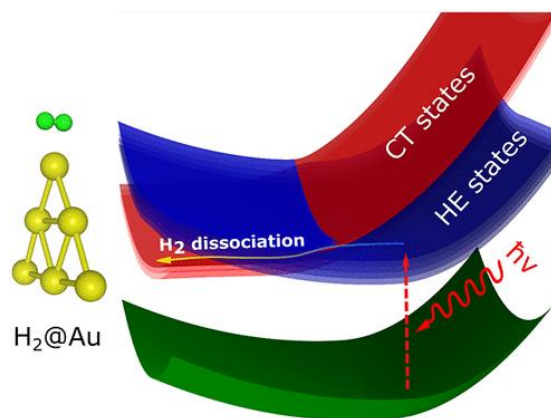
- [1] F. Plasser, *J. Chem. Phys.* **2020**, *152*, 084108.
- [2] P. Kimber, F. Plasser, *PCCP* **2020**, *22*, 6058–6080.
- [3] S. Mai, F. Plasser, J. Dorn, M. Fumanal, C. Daniel, L. González, *Coord. Chem. Rev.* **2018**, *361*, 74–97.
- [4] P. A. Sánchez-Murcia, J. J. Nogueira, F. Plasser, L. González, *Chem. Sci.* **2020**, *11*, 7685–7693.
- [5] F. Plasser *ChemPhotoChem* **2019**, *3*, 702.

Atomistic Understanding of Plasmon Mediated Photochemical Reactions

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Localized surface plasmon resonances (LSPRs) have attracted much recent attention for their potential in promoting chemical reactions with light. However, the mechanism of LSPR-induced chemical reactions is still not clear and suffers from many controversies. This presentation will discuss the atomic-scale mechanism of plasmonic hot-carrier mediated chemical reaction exemplified by H₂ dissociation by employing time-dependent density functional calculations theory and non-adiabatic molecular dynamics. The key observation is that there are nested excited states corresponding to both hot-electron excitation and charge transfer [1]. These nested states cross, facilitating the transitions depicted in the desorption induced by the electronic transitions model and the surface hopping model. I believe this is the first time that such a connection has been made based on a first-principles calculation. Diabatization of these states shows that the charge transfer states are responsible for H₂ dissociation, while the hot electron states do not. Previous works only identified the hot electron states, thus was unable to explain the dissociation in a convincing way. Moreover, we also found chemical reaction is tunable if the molecule is placed in the center of the plasmonic dimer [2]. The reaction rate can be either suppressed or enhanced depending on the geometry.

References:

- [1] Q. Wu, L. Zhou, G. C. Schatz, **Y. Zhang***, H. Guo*, *J. Am. Chem. Soc.* **2020**, 142, 13090–13101.
- [2] **Y. Zhang***, T. Nelson, S. Tretiak, H. Guo, G. C. Schatz. *ACS Nano*, **2018**, 12, 8415-8422.

How to connect

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 8

Time: Dec 17, 2020 09:30 AM Eastern Time (US and Canada)

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