SIMULATING PHOTOEXCITATION WITH A LASER PULSE BEYOND THE PERTURBATIVE LIMIT

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The advent of ultrashort laser pulses in the femtosecond to attosecond regime allows the study of ultrafast molecular dynamics with unprecedented time resolution [1,2]. These powerful modern light sources can result in the ionization of matter and thereby trigger electronic and nuclear dynamics [3,4]. In my talk, I will give an overview of the ongoing research efforts in the Worth group at UCL addressing the following fundamental questions: (i) Can we control photochemical processes by creating/manipulating a quantum superposition state with a laser pulse? (ii) Can we understand the coupled electron-nuclear motion and the associated ultrafast decoherence? (iii) Can we design laser pulses in a simple way to make use of the quantum interference pathways? (iv) Can we simulate an experimental photoelectron spectrum by developing simple theoretical models?

These elementary aspects of laser-matter interactions are governed by quantum mechanics and therefore we solve the time-dependent Schrödinger equation using state-of-the-art quantum dynamics method, MCTDH [5], in combination with vibronic coupling Hamiltonian [6]. This further allows us to deal with the non-adiabatic coupling between the electrons and the nuclei [6]. The ionized electron is modeled explicitly by incorporating the continuum of free-electron states [7]. The QUANTICS suite of programs are used to run the dynamical simulations [8].

References

[1] M. Nisoli, P. Decleva, F. Calegari, A. Palacios and F. Martín, Chem. Rev. 117, 10760 (2017).
[2] H. H. Fielding and G. A. Worth, Chem. Soc. Rev. 47, 309 (2018).
[3] V. Despré, N. V. Golubev and A. I. Kuleff, Phys. Rev. Lett. 121, 203002 (2018).
[4] A. Henley, J. W. Riley, B. Wang and H. H. Fielding, Faraday Discuss. 221, 202 (2020).
[5] M. H. Beck, A. Jäckle, G. A. Worth and H.-D. Meyer, Phys. Rep. 324, 1 (2000).
[6] G. A. Worth and L. S. Cederbaum, Annu. Rev. Phys. Chem. 55, 127 (2004).
[7] M. Seel and W. Domcke, J. Chem. Phys. 95, 7806 (1991).
[8] QUANTICS, http://www2.chem.ucl.ac.uk/quantics/doc/index.html

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