First-principles nonequilibrium Green's function approach to real- time simulations of correlated electrons in molecular systems

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Abstract

We show how to describe the correlated electron dynamics of molecules and molecular complexes driven by ultrafast laser pulses using the first-principles nonequilibrium Green's function (NEGF) method [1-5]. The method is currently implemented in the CHEERS code [6] which allows for addressing a broad variety of time-resolved phenomena such as transient photoabsorption and photoemission, time-resolved Auger decays as well as ultrafast charge migration. CHEERS is compatible with all main single-particle basis functions (plane-waves, Gaussian type orbitals, Slater type orbitals, real-space grid etc.) and it accounts for both static and dynamical correlation effects.

In this talk we discuss how the proposed method is used to address the charge migration process in biological molecules [7], a topic of high current interest to unravel the fundamental mechanisms at the basis of photo-protection or photo-damage. The role played by correlations and memory effects is crucial to achieve an excellent agreement with available experimental data [7-9].

References:

[1] S. Latini, E. Perfetto, A.M. Uimonen, R. van Leeuwen and G. Stefanucci, Phys Rev. B 89, 075306 (2014).
[2] E. Perfetto, A.M. Uimonen, R. van Leeuwen and G. Stefanucci, Phys. Rev. A 92, 033419 (2015).
[3] F. Covito, E. Perfetto, A. Rubio and G. Stefanucci, Phys. Rev. A 97, 061401(R) (2018).
[4] D. Karlsson, R. van Leeuwen, E. Perfetto and G. Stefanucci, Phys. Rev. B 98, 115148 (2018).
[5] E. V. Boström, A. Mikkelsen, C. Verdozzi, E. Perfetto and G. Stefanucci, Nano Lett. 18, 785 (2018).
[6] E. Perfetto and G. Stefanucci, Journal of Physics: Condensed Matter 30, 465901 (2018).
[7] E. Perfetto, D. Sangalli, A. Marini and G. Stefanucci, J. Phys. Chem. Lett. 9, 1353 (2018).
[8] F. Calegari et al, Science 346, 336 (2014).
[9] Lara-Astiaso et al, J. Phys. Chem. Lett. 9, 4570 (2018)