Extending the tool set for excited states in solids — from static excited-state properties to non-adiabatic molecular dynamics

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In recent years, significant milestones have been achieved for the description of excited states in molecular systems using non-adiabatic molecular dynamics, while the development of analogous ansätze for extended systems implying periodic boundary conditions is still lagging behind. With the aim to create an equally extensive tool set for time-dependent density functional theory (TDDFT) methods based on the Gaussian and plane waves (GPW) method [1,2,3], we will present recent developments, focusing on the interface of CP2K [4] with NewtonX [5] as well as on on-going developments to go beyond conventional TDDFT. Benchmark results will demonstrate applicability at the large scale as well as straight-forward comparison and validation with respect to state-of-the-art program packages such as Turbomole [6] and Octopus [7].

[1] A. Hehn, B. Sertcan, F. Belleflamme, S. K. Chulkov, M. B. Watkins, J. Hutter, *J. Chem. Theory Comput. 18*, 4186 (**2022**).

[2] B. Sertcan, J. Hutter, A. Hehn, *J. Chem. Theory Comput. J. Chem. Theory Comput.*, 20, 8494 (**2024**).

[3] J.-R. Vogt, J. Wilhelm, A. Hehn, *J. Chem. Phys. 162*, 082502 (**2025**).

[4] T. Kühne *et al.*, *J. Chem. Phys. 152*, 194103 (**2020**).

[5] M. Barbatti *et al.*, *J. Chem. Theory Comput. 18*, 6851 (**2022**).

[6] Y. J. Franzke *et al.*, *J. Chem. Theory Comput. 19*, 6859 (**2023**).

[7] A. Castro *et al.*, *Phys. Stat. Sol. (b) 243*, 2465 (**2006**).