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Semiclassical Frozen Gaussian Propagation Approach for Molecular Dynamics with Nonadiabatic Transitions

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The modern tends in physics of molecular interactions in chemical and biological systems, polyatomic molecule – laser interactions and further development of optimal control theory (OCT) needs development of effective semiclassical approaches for wave packet propagation with nonadiabatic transition. Natural treatment of various quantum effects becomes an important requirement for such theories.

In the present work electronically nonadiabatic photodissociation process is considered. In the adiabatic representation the whole process can be divided into three steps: (i) propagation on an initial potential, (ii) nonadiabatic transition, and (iii) propagation on a final potential. Semiclassical approach developed naturally combines the advantages of the most effective methods: Herman-Kluk method [1] for single surface propagation at (i), (iii) and semiclassical Zhu-Nakamura theory [2, 3] for nonadiabatic transition (ii).

The theory is based on natural mathematical principles that straightforwardly lead to quite simple formulas for trajectory propagator and probabilities of nonadiabatic passing, reflection and hopping without any additional physical assumptions.

The results of testing calculations for 1D (H_2^+ in laser field) and 2D (HOD in laser field) systems are presented. The theory was found to work well for propagation durations of several molecule vibrational periods and wide wavepacket energy ranges.

References

[1] See, B. R. McQuarrie and P. Brumer, Chem. Phys. Lett. 319, 27 (2000), and references therein.
[2] C. Zhu, Y. Teranishi, and H. Nakamura, Adv. Chem. Phys. 117, 127 (2001).
[3] H. Nakamura, Nonadiabatic transition: Concepts, Basic Theories and Applications, World Scientific, Singapore (2002).