Femtosecond wave packet dynamics and vibrational decoherence: a mixed quantum-classical study of I_2 in a dense rare gas environment

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An important aspect of quantum dynamics in an environment is ultrafast vibrational relaxation. Additionally, if several initial vibrational states are excited coherently, the system-solute interaction not only leads to energy relaxation but also to decoherence.
The system under consideration is the I_2 molecule in a dense rare gas environment. The excitation by femtosecond laser pulses and the subsequent relaxation dynamics is studied using quantum and mixed quantum/classical methods.

In a first part, quantum effects that rely on a phase coherent temporal evolution like wave packet revivals or interferences are presented. Specifically, it is shown how wave packet interferences should be measurable in a pump-probe set-up, a result that is currently being verified experimentally.
In a second part, we show the effect of random rare gas collisions onto the coherent wave packet motion. To this end, the quantum wave packet propagation is combined with a classical molecular dynamics simulation. The good agreement with a series of pump-probe experiments performed by the Zewail group confirms the validity of our theoretical modelling and allows for a clear interpretation of the observed structures in terms of decoherence processes.