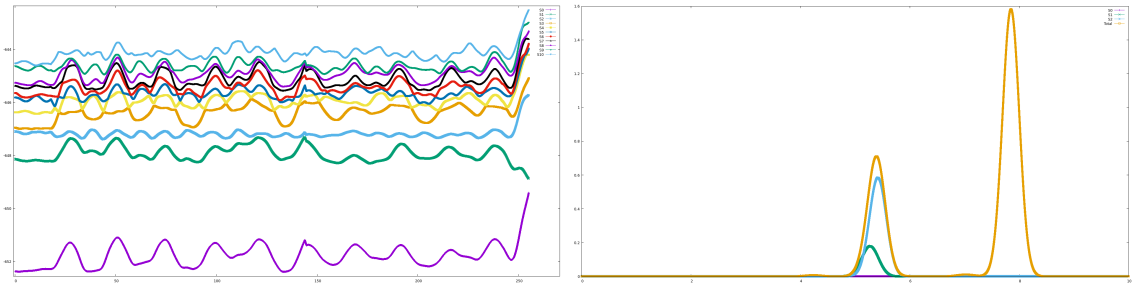
Report

of Jakub Martinka

Excited States and Nonadiabatic Dynamics CyberTraining Workshop 2021

For my project, I chose vinyl chloride as a test molecule. Vinyl chloride is mainly used for the preparation of polyvinyl chloride (PVC). At first, I wanted to use vinyl bromide as a test molecule, but NEXMD does not support the Br atom yet. Because of that, I used a similar molecule - vinyl chloride. The reason for choosing this molecule was that I have already worked with a similar one. It is a small molecule, making it an ideal system for test trajectories because in the case of very accurate methods it is a good idea not to perform expensive calculations. I also chose this molecule because I will be working on machine-learning of spin-orbital couplings in the future and I will use vinyl bromide as a test molecule. I was interested in using various programs to study this molecule, especially I focused on NEXMD and NewtonX using DFTB+, I also tried QXMD but after several problems, I gave up. I followed the NEXMD outline in the standard way that the manual recommends, so I got, for example, Figure 1a and the spectrum in Figure 1b, but I did not further analyze obtained information.



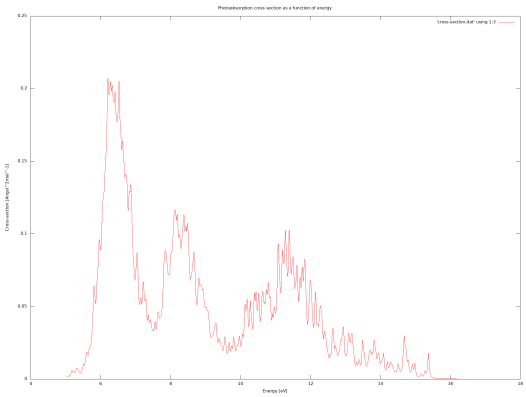
(a) Potential energy as a function of time

(b) Absorption spectra

Figure 1: Results for NEXMD

The main part of my project was the use of Newton X by FSSH-NAMD with DFTB+, for example for spectrum simulation. I followed the standard procedure, which is also described in the tutorial, I prepared the structure and optimized it, calculated the normal modes, and used it to create 500 initial conditions. I used the halorg kit, which I downloaded from the official dftb+ website. Using NewtonX, I created a spectrum (Figure 2a)that is similar in the peak position compared to the experimental spectrum Figure 2b. Then I prepared the input for SH-NAMD. I selected the initial state according to the oscillator strenght as S_2. Then I started a total of 30 trajectories. Most of them terminated around 60 fs. I analyzed the trajectory using available statistical methods, the results of which I present below.

The pictures show the potential energies of the ten states as a function of time. The blue triangles indicate the current state of the system at each time step. Figure 3 shows a fraction of trajectories and average adiabatic population in the second excited state as a function of time.



(a) Photoabsorption cross section ($\mathring{A}^2$.molecule^{-1}) as a function of energy (eV).

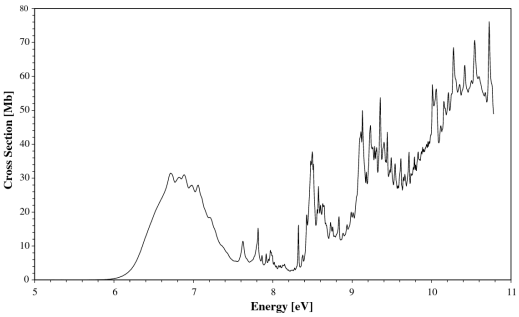


Fig. 1. The VUV photo-absorption spectrum of C_2H_3Cl in the 5.0–11.0 eV energy region.

(b) Experimental spectra (Limão-Vieira *et. al.*, CP 2006)

Figure 2: Potential energies of some trajectories - ten states as a function of time. The blue triangles indicate the current state of the system at each time step.

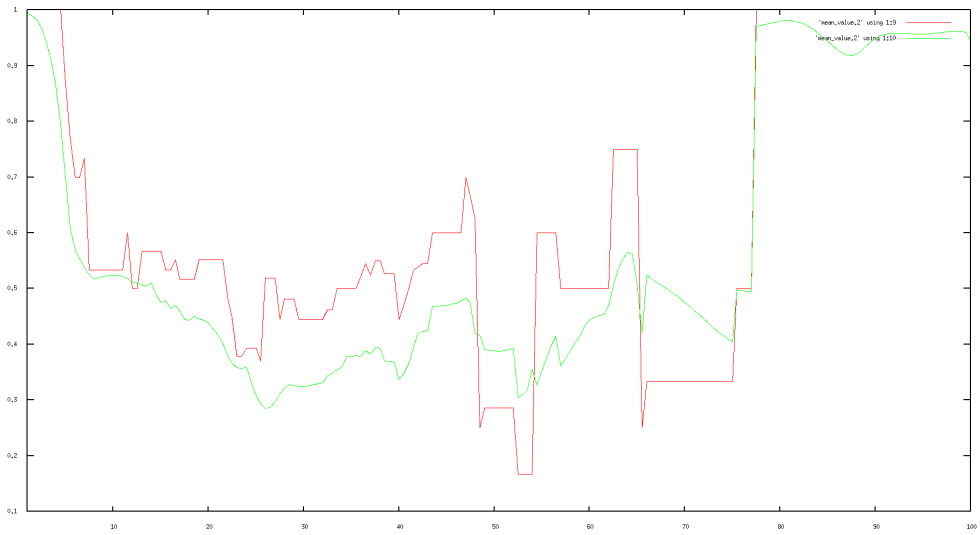
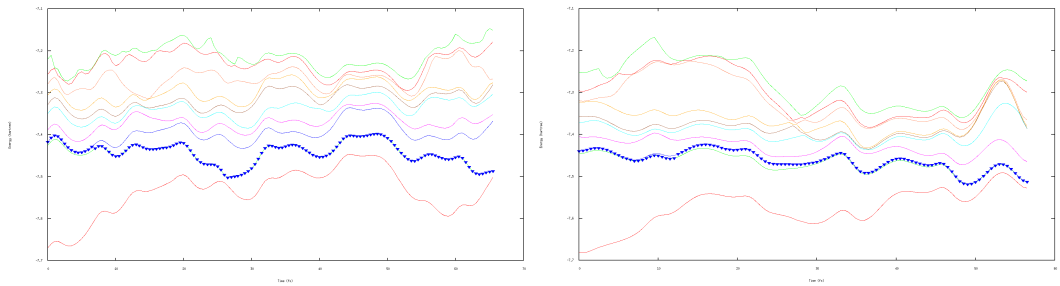
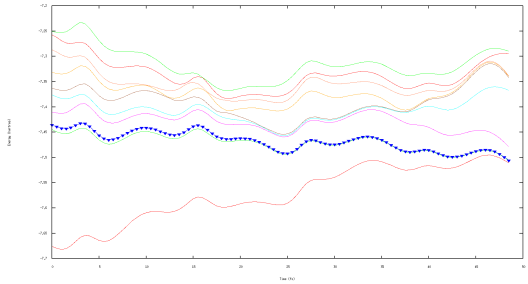


Figure 3: Fraction of trajectories and average adiabatic population in the second excited state as a function of time

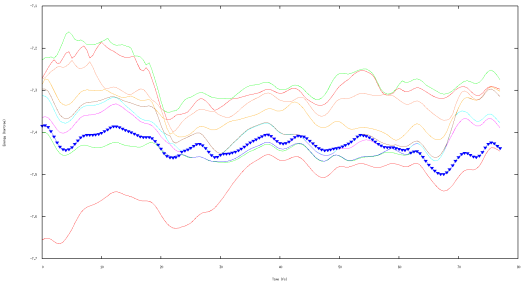


(a) TRAJ36

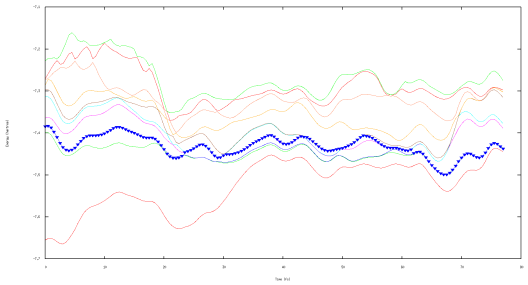
(b) TRAJ37



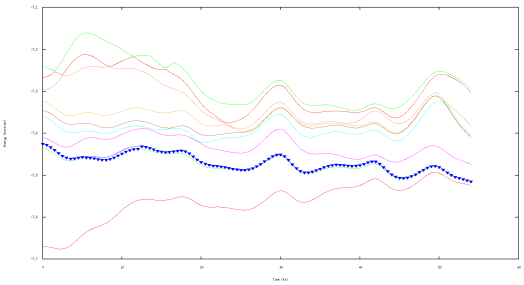
(c) TRAJ42



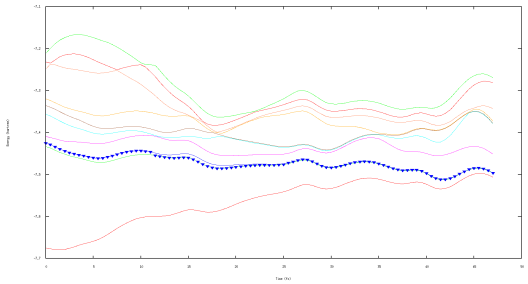
(d) TRAJ45



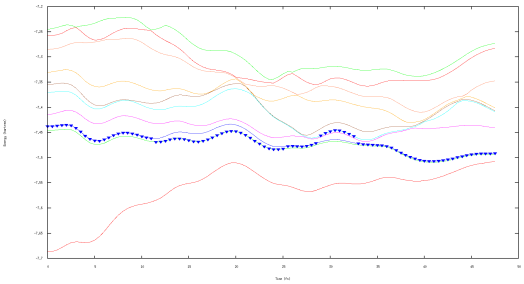
(e) TRAJ48



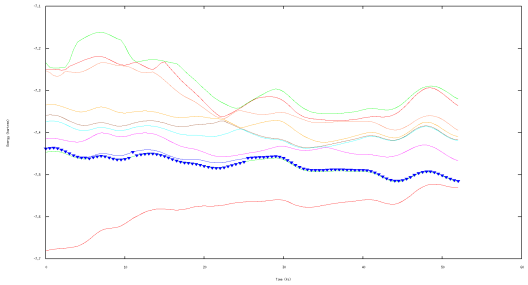
(f) TRAJ49



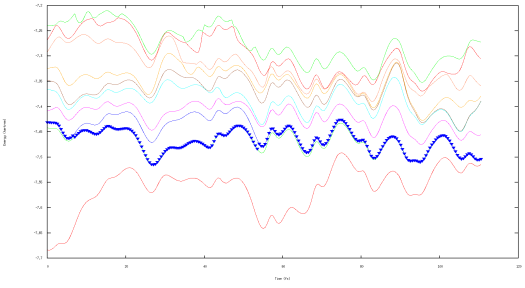
(g) TRAJ50



(h) TRAJ52



(i) TRAJ57



(j) TRAJ59

Figure 4: Potential energies of some trajectories - ten states as a function of time. The blue triangles indicate the current state of the system at each time step.