

1. File Fig3.tar.gz. Contains:

1.1. File Fig3A.eps - Figure 3A in postscript format.

1.2. File Fig3A.fig - Figure 3A in Matlab format.

1.3. File Fig3A_data - data for Figure 3A

Column 1 - time in fs

Column 2-4 - populations of electronic states (arb. units)

1.4. Files Fig3B.eps, Fig3B.fig and Fig3B_data -the same as above for Figure 3B

2. File Fig4.tar.gz. Contains:

2.1. File Fig4.eps - Figure 4 in postscript format.

2.2. File Fig4.fig - Figure 4 in Matlab format.

2.3. File Fig4_Data_Calc - calculated TKER spectrum of pyrrole:

Column 1 - energy in cm⁻¹

Columns 2-3 - TKER spectra for two different wavelengths (see paper for the details)

2.4. Files Fig4_Data_Exp_2 and Fig4_Data_Exp_3 - experimental TKER spectra of pyrrole for two different wavelengths

Column 1 - energy in cm⁻¹

Column 2 - TKER spectrum

3. Files "traj_20.tar.gz" - "traj_29.tar.gz" contain raw trajectory data for 200 initial geometries randomly generated from Wigner distribution. Each file contains data for 10 initial geometries.

Directories pyrrol_g_100 - pyrrol_g_299 correspond to the above random geometries. Each of them contains a parent ground state trajectory and subdirectories freq_20 and freq_30 for two wavelengths. These subdirectories contain subdirectories for child trajectories in format clon_XXX_Y, where XXX is a step where the child trajectory was cloned out of the parent trajectory, and Y is its initial electronic state. They contain, in turn, subdirectories tp1 and tp2. When only tp1 subdirectory is present - continuation of calculations. When both tp1 and tp2 subdirectories are present - branching at a cloning point: each of subdirectories contains one branch.

All trajectory data (including initial geometries) are written in files "Trajectory", which are ascii files with obvious format.

4. File TKER_raw_data.tar.gz - TKER_2 and TKER_3 contain raw TKER data for two different wavelengths

Column 1 - initial geometry number

Column 2 - branch number

Column 3 - branch weight

Column 4 - N-H distance in Å

Column 5 - H atom kinetic energy in cm⁻¹

Columns 6-8 - H atom momentum in a.u.

5. File "pyrol_k_2000.com" - input file for MOLPRO.

This file is the same for all calculations, as a geometry is defined by a separate file.