1. File 'fig1.tar.gz' contains data for Figure 1.

1.1 File 'fig1a.eps' - Figure 1a in postscript format.
1.2 File 'fig1b.eps' - Figure 1b in postscript format.
1.3 Files experiment_TKER_***.csv - data for the experimental portion of Figures 1a and 1b where *** is the pump wavelength used. The deuterated derivative is given the label '2-EPD' in the filename.
    Column 1 - energy in cm-1
    Column 2 - intensity of TKER spectrum
1.4 Files AIMC_TKER_***.csv - data for the ab initio multiple cloning calculated portions of Figures 1a and 1b where *** is the calculation identifier (2-EP_S1=2-EP(S1), 2-EP_S2=2-EP(S2), or 2-EPD=2-EP-d1(S1))) as labelled in the paper.
    Column 1 - energy in cm-1
    Column 2 - intensity of TKER spectrum

2. File 'fig2.tar.gz' contains data for Figure 2.

2.1 File 'fig2a.eps' - Figure 2a in postscript format.
2.2 File 'fig2b.eps' - Figure 2b in postscript format.
2.3 Files 'fig2c_axis1.eps' and 'fig2c_axis2.eps' - both parts of Figure 2c in postscript format.
2.4 File 'fig2d.eps' - Figure 2d in postscript format.
2.5 File 'exp_262_VMI.csv' - data for Figure 2a, the experimental velocity map image as a matrix.
2.6 File 'AIMC_VMI.csv' - data for Figure 2b, the ab initio multiple cloning calculated velcoity map image, given in polar coordinates as defined by Equation (2) in the text
    Column 1 - modulus of velocity, r in polar coordinates from Equation (2) in main text, in units of A fs-1
    Column 2 - angle, phi in polar coordinates from Equation (2) in main text
    Column 3 - intensity
2.7 File 'velocity_data_2-EP_S1.csv' - data for Figure 2d
    Column 1 - velocity in the x direction in A fs-1
    Column 2 - velocity in the y direction in A fs-1
    Column 3 - velocity in the z direction in A fs-1
    Column 4 - density of velocity distribution at that point

3. File 'fig3.tar.gz' contains data for Figure 3.

3.1 File 'fig3a.eps' - Figure 3a in postscript format.
3.2 File 'fig3b.eps' - Figure 3b in postscript format.
3.3 File 'fig3c.eps' - Figure 3c in postscript format.
3.4 Files 'AIMC_smoothed_***.csv' - smoothed data from the ab initio multiple cloning calculations for Figures 3a, b and c. *** is the calculation identifier (2-EP_S1=2-EP(S1), 2-EP_S2=2-EP(S2), or 2-EPD=2-EP-d1(S1))) as labelled in the paper.
    Column 1 - time in fs
    Column 2 - normalised H/D dissociation count
3.5 Files 'AIMC_raw_***.csv' - raw data from the ab initio multiple cloning calculations for Figures 3a, b and c. *** is the calculation identifier (2-EP_S1=2-EP(S1), 2-EP_S2=2-EP(S2), or 2-EPD=2-EP-d1(S1))) as labelled in the paper.
    Column 1 - dissociation time in fs
    Column 2 - normalised cumulative sum of dissociation weight
3.6 Files 'experiment_kinetic_lifetime_***.csv' - data for the experimental H/D appearance lifetimes from Figures 3a, b and c. *** is the pump wavelength used. The deuterated derivative is given the label '2-EPD' in the filename.
    Column 1 - time in fs
    Column 2 - normalised H/D dissociation count
3.7 File 'plot_figure3a.gnu' - example of the gnuplot file used to plot Figures 3a, b and c, showing the fitting function used on the smoothed data from the ab initio multiple cloning calculations

4. File 'fig4.tar.gz' contains data for Figure 4

4.1 File 'fig4.eps' - Figure 4 in postscript format.
4.2 File 'AIMC_pops_2-EP_S2.csv' - data for Figure 4, averaged electronic state populations for all 2-EP(S2) trajectories

    Column 1 - time in fs
    Column 2 - population of S0 electronic state
    Column 3 - population of S1 electronic state
    Column 4 - population of S2 electronic state

5. File 'fig_s1.tar.gz' contains data for Figure S1 from the ESI

5.1 File 'fig_s1.eps' - Figure S1 in postscript format
5.2 File '2-EP_S1_diss_time_init_energy.csv' - data for Figure S1
    Column 1 - dissociation time in fs
    Column 2 - initial energy in a.u.

6. File 'fig_s2.tar.gz' contains data for Figure S2 from the ESI

6.1 File 'fig_s2a.eps' - Figure S2a in postscript format
6.2 File 'fig_s2b.eps' - Figure S2b in postscript format
6.3 File '2-ep_pe_static_fast.tab' - Data for the static potential energy curves of Figure 2a
    Column 1 - N-H bond length in Angstroms
    Column 2 - S0 electronic state energy in a.u.
    Column 3 - S1 electronic state energy in a.u.
    Column 4 - S2 electronic state energy in a.u.
6.4 File '2-EP_PE_dynamics_fast.csv' - Data for the dynamic potential energy of the trajectory shown in Figure 2a
    Column 1 - N-H bond length in Angstroms
    Column 2 - Trajectory potential energy in a.u.
6.5 File '2-ep_pe_static_slow.tab' - Data for the static potential energy curves of Figure 2b
    Column 1 - N-H bond length in Angstroms
    Column 2 - S0 electronic state energy in a.u.
    Column 3 - S1 electronic state energy in a.u.
    Column 4 - S2 electronic state energy in a.u.
6.6 File '2-EP_PE_dynamics_slow.csv' - Data for the dynamic potential energy of the trajectory shown in Figure 2b
    Column 1 - N-H bond length in Angstroms
    Column 2 - Trajectory potential energy in a.u.

7. Files '2-EP_S1_1_traj.tar.gz' and '2-EP_S1_2_traj.tar.gz' contains data for all 600 2-EP(S1) trajectories with individual initial geometries, split into two sets of 300 trajectories. Each trajectory has its own directory which is assigned a number from 100-699. These contain subdirectories tp1 and tp2. When directory tp1 is present, it is a continuation of the calculation. When directory tp2 is present there has been a cloning event, and the directory contains a branch of the trajectory. All trajectory data is written in the file "Trajectory" which is present in each directory, written in ASCII format with appropriate headings for the data.

8. Files '2-EP_S2_1_traj.tar.gz' and '2-EP_S2_2_traj.tar.gz' contains data for all 600 2-EP(S2) trajectories with individual initial geometries, split into two sets of 300 trajectories. The structure is as above.

9. Files '2-EPD_1_traj.tar.gz' and '2-EPD_2_traj.tar.gz' contains data for all 600 2-EP-d1(S1) trajectories with individual initial geometries, split into two sets of 300 trajectories. The structure is as above.

10. File '2-EP.com' contains an example MOLPRO input file for the calculation, which is the same for all geometries as the geometry is read by a separate file.