Supporting Information for

"Dependence of Adenine Raman Spectrum on Excitation Laser Wavelength: Comparison between Experiment and Theoretical Simulations"

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The experimental detail, comparison of the powder and aqueous Raman spectra, comparison of the calculated peak position, Raman activity ratio versus excitation laser wavenumber for different calculation methods, schematic vibrational modes of the adenine molecule, hydrogen bonded dimer,π-stacked dimer, and hydrogen bonded trimer, the numerical results from the sum over state formula, as well as the XYZ geometry of the calculated structures are given.

1. Experimental Details

The white-light calibration lamp system used in this study is the NIST-traceable DH-2000 from Ocean Optics, which contains deuterium and halogen lamps. The spectral irradiance is shown below. Since the wavelength range of interest is longer than 500 nm, only the halogen lamp was used in the spectral intensity calibration.

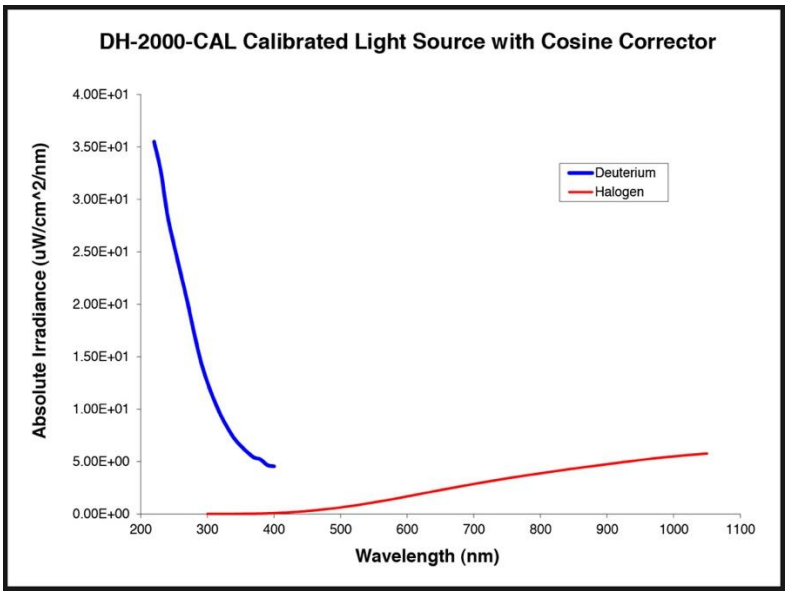


Figure S1. The spectral irradiance of the DH-2000 white-light calibration lamp.

Below we present the variation of the Raman spectra as we perform the intensity calibration.

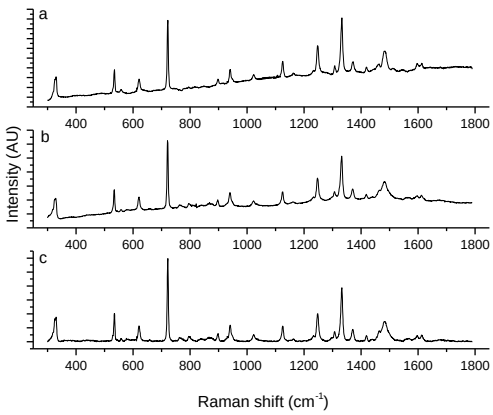


Figure S2. The Raman spectra of adenine powder obtained using 532 nm excitation laser: (a) before intensity calibration, (b) after intensity calibration, and (c) after subtraction of baseline from (b).

2. Direct Comparison of the Aqueous and Powder Raman spectra

As can be seen below, slight differences in intensity can be seen between the Raman spectra in the two phases, but the peak positions do not show much variation.

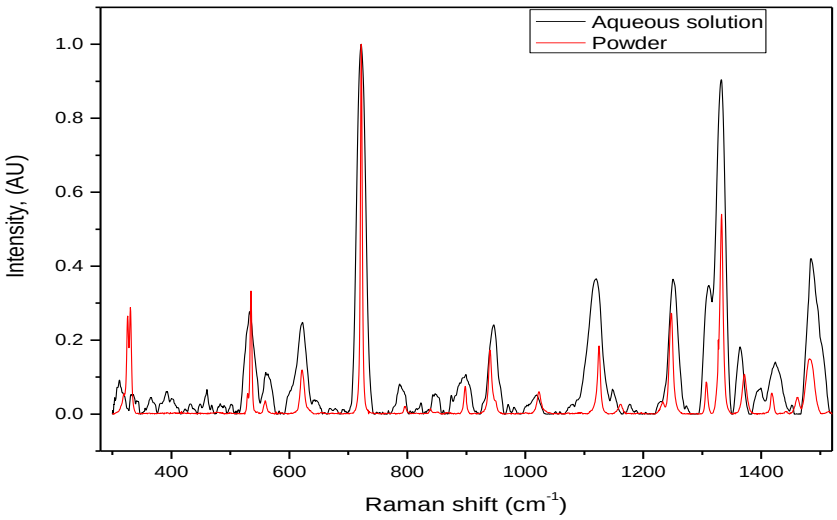


Figure S3. Raman spectra of adenine in aqueous solution and powder phase.

3. Vibrational calculation results

Below we compare the peak positions obtained experimentally with those obtained from various density functional theory (DFT) functionals.

Table S1. The experimental and calculated vibrational peak positions, in cm^{-1}, for adenine studied in the present paper.

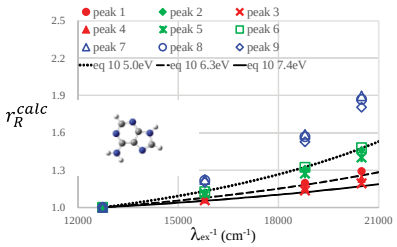
Peak	1	2	3	4	5	6	7	8	9
Experimental powder	534	621	722	940	1023	1125	1247	1332	1482
Experimental aqueous sol	533	621	722	946	1019	1120	1250	1332	1484
B3LYP monomer	520	618	725	945	1008	1079	1242	1356	1513
B3LYP-D2 monomer	522	618	724	942	1014	1068	1242	1354	1511
B3LYP-D2 H-bond dimer	534	629	727	951	1043	1076	1261	1356	1517
B3LYP-D2 π-stacked dimer	528	621	726	944	1041	1074	1255	1355	1515
B3LYP-D2 H-bond trimer	536	637	728	949	1056	1073	1264	1356	1505
B3LYP/AVTZ monomer	520	618	726	945	1009	1080	1242	1355	1514
B3PW91 monomer	521	619	731	945	1012	1091	1273	1380	1529
M06-2X monomer	535	625	737	954	1022	1094	1281	1390	1555
B3LYP monomer water	532	620	724	951	1012	1098	1237	1356	1515
B3LYP monomer acetone	520	620	724	946	1005	1090	1236	1356	1508
B3LYP monomer benzene	518	620	724	946	1007	1090	1236	1356	1508

Table S2. Root mean square error δ, in cm^{-1}, of the 9 calculated peak position for adenine. We compared to the powder phase experimental results. Other than B3LYP/AVTZ which used aug-cc-pVTZ basis set all calculations were done with 6-311+G(d,p) basis set.

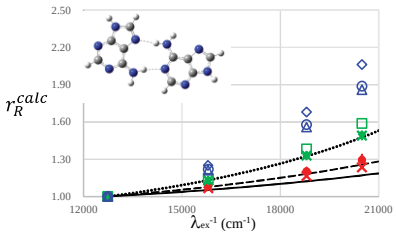
Method	Error δ
B3LYP monomer	23
B3LYP-D monomer	25
B3LYP-D H-bond dimer	25
B3LYP-D π-stacked dimer	24
B3LYP-D H-bond trimer	26
B3LYP/AVTZ monomer	22
B3PW91 monomer	29
M06-2X monomer	38
B3LYP monomer SMD water	18
B3LYP monomer SMD acetone	20
B3LYP monomer SMD benzene	20

4. Raman activity ratio as a function of excitation laser wavenumber

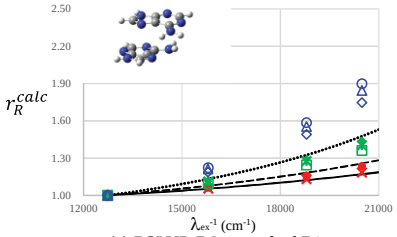
In Figure S4, we present the comparison of the excitation laser wavenumber dependence of the Raman activity ratio calculated for adenine molecule and clusters. Next the density functional theory functional dependence is given in Figure S5. All calculations show that the excitation laser wavenumber dependence is stronger for the peaks 7, 8, and 9 compared to peaks 1, 3 and 4. This general trend does not depend on the hydrogen bonding or π-stacking interactions, as seen for Figure S4. One difference may be that for peak 9 in Figure S4, the intensity increase with increase in excitation laser wavenumber is greater when it is hydrogen bonded in the dimer and trimer state compared to the bare monomer case. Figure S5 shows that the change of B3LYP to B3PW91 or M06-2X does not change the trend much. The use of larger basis set, B3LYP/aug-cc-pVTZ, does not cause much change in the peak position compared to the obtained from B3LYP/6-311+G(d,p) (see Table 1). Furthermore, it also gives similar excitation laser wavelength dependence for the Raman activity other than for peak 1 (see Figure S5).



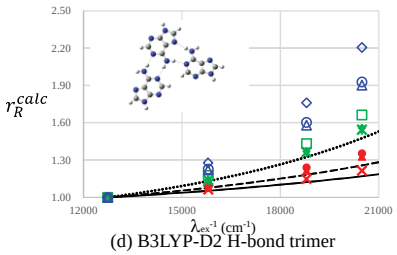
(a) B3LYP Monomer



(b) B3LYP-D2 H-bond Dimer

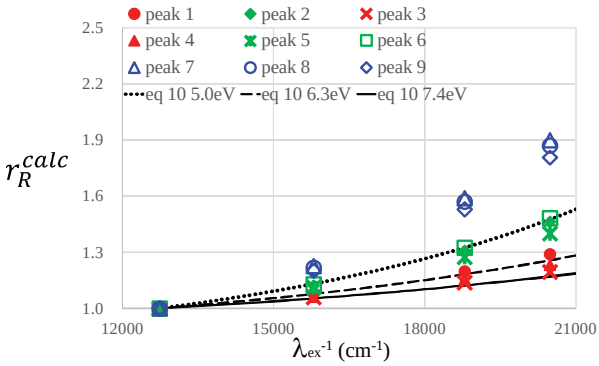


(c) B3LYP-D2 p-stacked Dimer

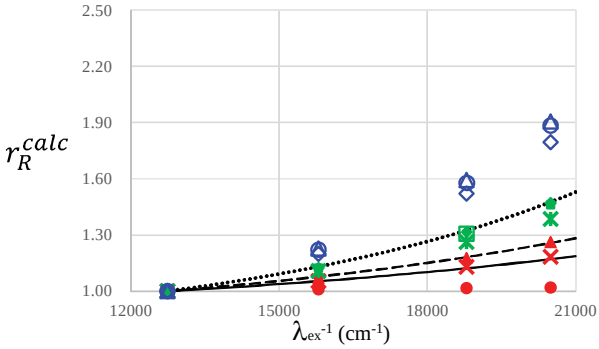


(d) B3LYP-D2 H-bond trimer

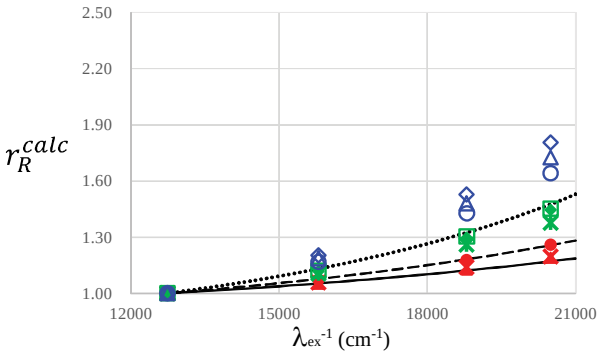
Figure S4. Raman activity ratio r_R^{calc} of adenine dimer and trimer of 9 Raman peaks obtained with B3LYP-D2/6-311+G(d,p), as well as, $r_R(\lambda_{ex}, \Omega_j, k')$ obtained by eq 13 for three lowest energy absorption signatures of adenines at 5.0, 6.3 and 7.4 eV.



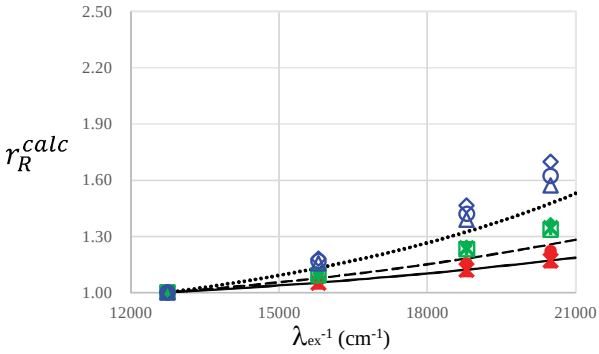
(a) B3LYP/6-311+G(d,p)



(b) B3LYP/aug-cc-pVTZ



(c) B3PW91/6-311+G(d,p)



(d) M06-2X/6-311+G(d,p)

Figure S5. Raman activity ratio r_R^{calc} of adenine monomer of 9 prominent Raman peaks obtained with B3LYP/aug-cc-pVTZ, B3PW91/6-311+G(d,p) and M06-2X/6-311+G(d,p), as well as, $r_R(\lambda_{ex}, \Omega_j, k')$ obtained by eq 13 for three lowest energy absorption signatures of adenines at 5.0, 6.3 and 7.4 eV *vs*. excitation photon energy E_{ex}.

5. Vibrational modes of adenine molecule, dimer and trimer

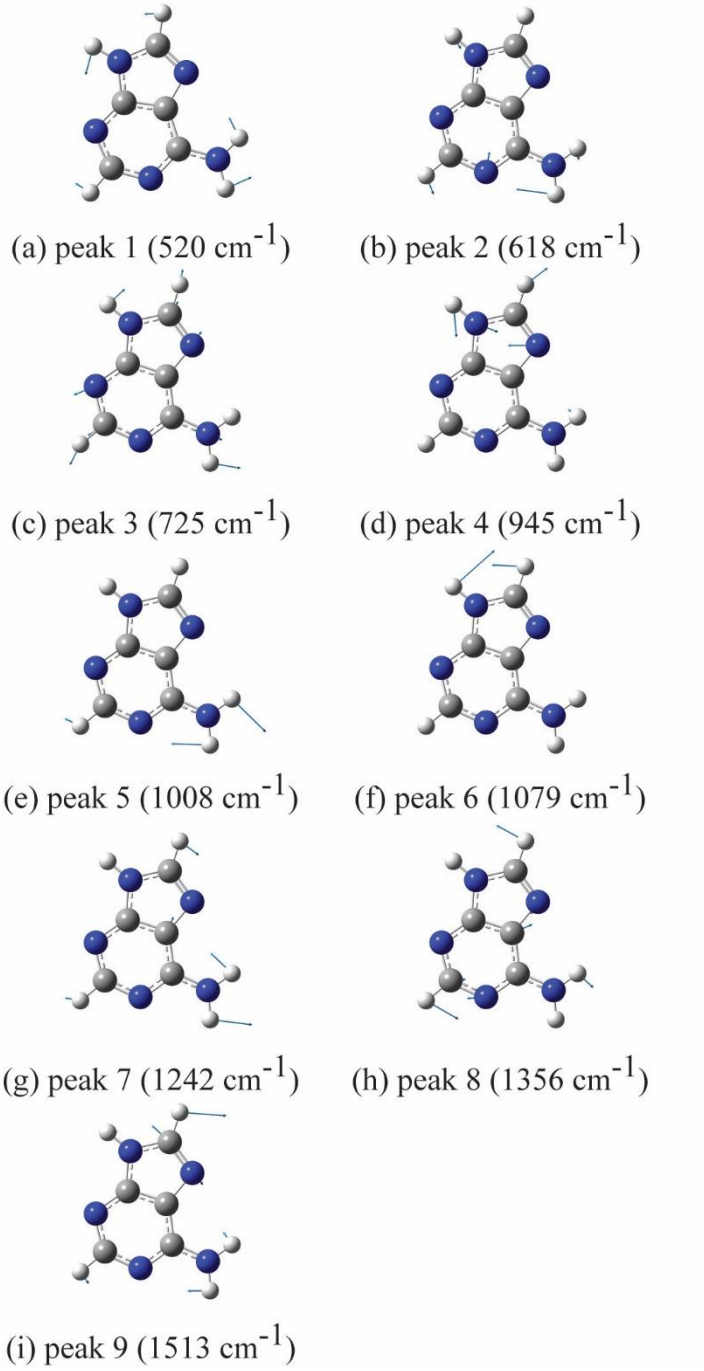


Figure S6. Vibrational modes for the adenine molecule

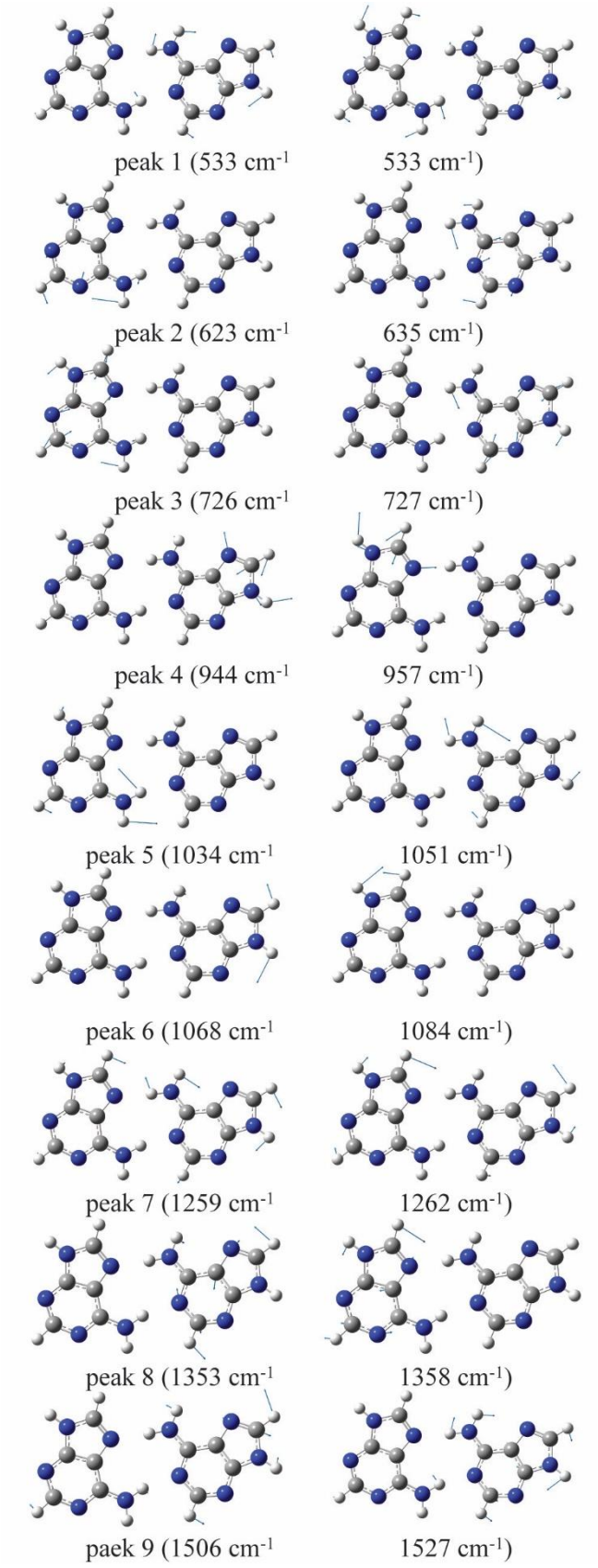


Figure S7. Vibrational modes for the hydrogen bonded dimer

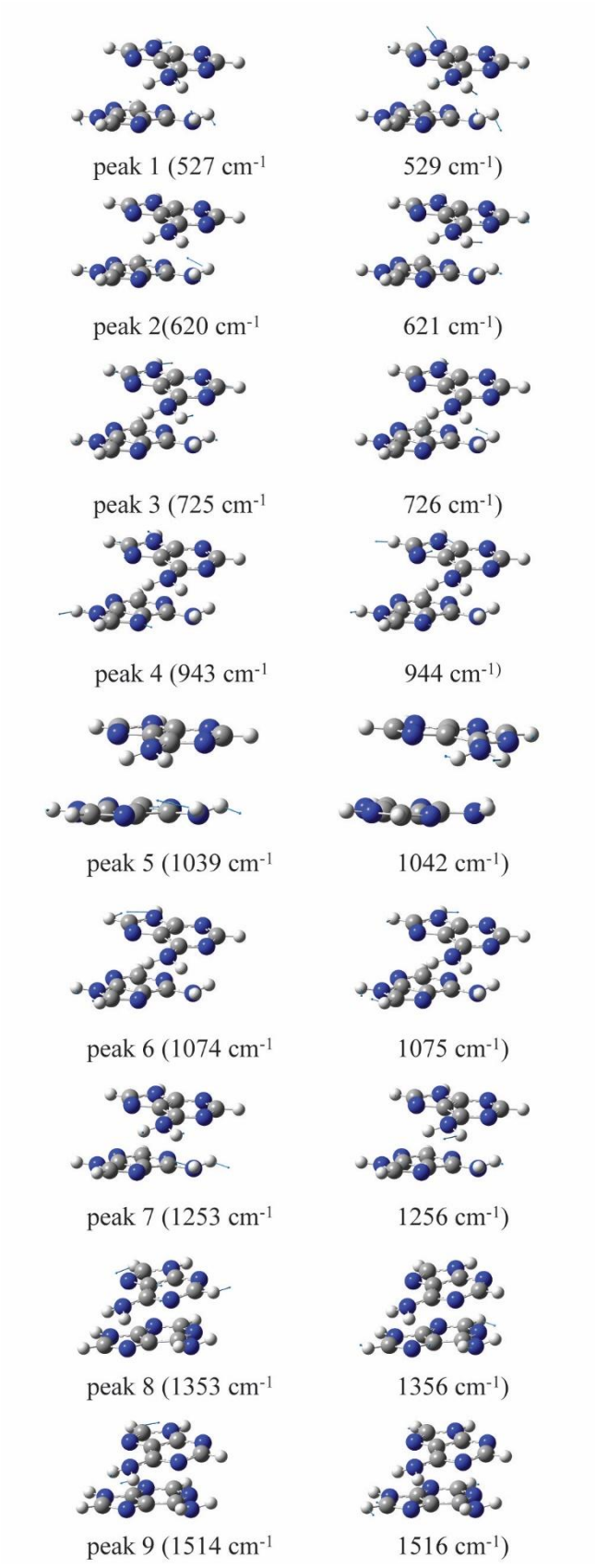


Figure S8. Vibrational modes for the π-stacked dimer

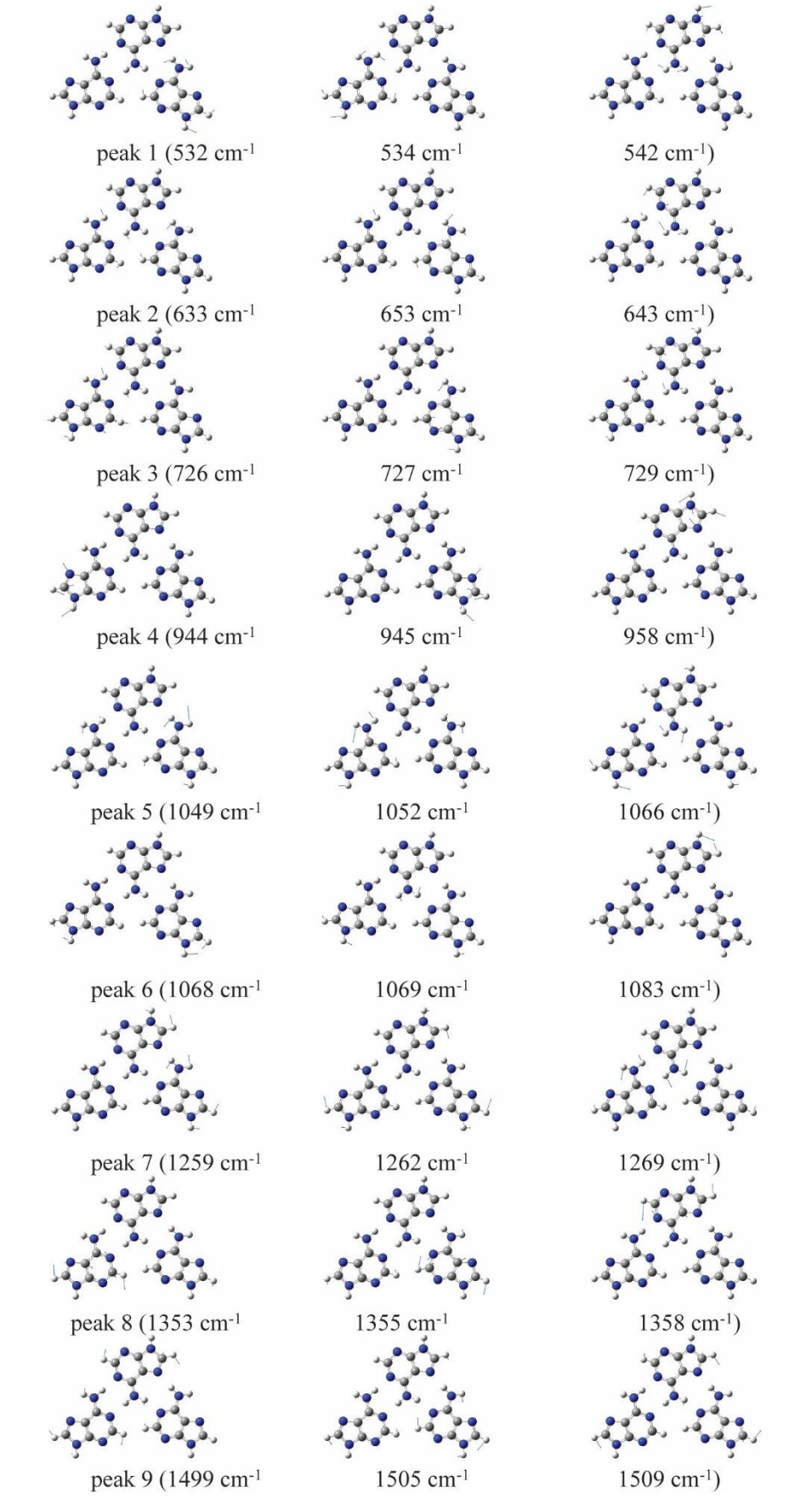


Figure S9. Vibrational modes for the hydrogen bonded trimer

Sum of state formulation of the excitation wavelength dependent polarizability

As written in the main text the Raman activity is given from the normal mode derivative of the excitation laser wavelength dependent polarizability:

$$\frac{\partial \alpha_{\gamma\sigma}(\lambda_{\mathrm{ex}})}{\partial Q_j} = 2\sum_{k\neq 0} \frac{E_k - E_0}{(E_k - E_0)^2 - \left(\frac{hc}{\lambda_{ex}}\right)^2} \frac{\partial}{\partial Q_j}\left(Re\left[\langle 0|\mu_\gamma|k\rangle\langle k|\mu_\sigma|0\rangle\right]\right)$$

In the following Tables S3-S11 we will list the values for each k[th] electronic state in the sum, $\frac{E_k - E_0}{(E_k - E_0)^2 - \left(\frac{hc}{\lambda_{ex}}\right)^2} \frac{\partial}{\partial Q_j}\left(Re\left[\langle 0|\mu_\gamma|k\rangle\langle k|\mu_\sigma|0\rangle\right]\right)$, for the first 99 excitations obtained from the TD-B3LYP/6-311+G(d,p) for the adenine monomer. As can be seen in Tables S3-S11, large contributions to the derivative of the polarizabilty is seen from states near 6.25 eV and 7.39 eV, however the values have plus and minus contributions that cancel each other. Furthermore, we notice that the contribution is coming from many different electronic states, and we are not able to assign the normal mode derivative of the polarizability as coming from one certain electronic excited state.

Table S3. Contribution of the k[th] excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 1. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	-0.02	0.00
4.989	-0.41	0.23	0.00	-1.01
5.253	0.91	0.65	0.00	0.85
5.395	0.00	0.00	-0.06	0.00
5.555	0.00	0.00	0.00	0.00
5.660	0.00	0.00	0.09	0.00
5.852	0.00	0.00	0.00	0.00
6.169	-0.44	-7.75	0.00	2.32
6.206	0.01	3.50	0.00	-0.17
6.253	0.12	4.92	0.00	-2.37
6.254	0.00	0.00	0.00	0.00
6.506	0.00	0.00	0.00	0.00
6.560	0.00	0.00	-0.10	0.00
6.572	0.14	0.00	0.00	0.06
6.595	0.00	0.00	0.51	0.00
6.611	0.00	0.00	-1.45	0.00
6.617	0.00	0.00	1.03	0.00
6.646	0.00	-1.46	0.00	-0.46
6.869	0.00	0.00	-0.06	0.00
6.888	-0.06	0.06	0.00	0.05
6.959	0.00	0.00	0.00	0.00
7.014	0.09	-0.21	0.00	-0.24
7.072	-2.12	0.21	0.00	0.84
7.133	0.00	0.00	0.00	0.00
7.179	0.17	0.06	0.00	0.11

7.312	0.81	-0.71	0.00	-1.06
7.332	0.00	0.00	0.01	0.00
7.370	0.86	3.91	0.00	-2.29
7.398	4.73	-0.62	0.00	-2.61
7.406	-4.12	-3.18	0.00	5.82
7.477	0.00	0.00	-0.01	0.00
7.509	0.18	0.41	0.00	-0.29
7.537	0.00	0.00	-0.02	0.00
7.564	0.00	0.00	-0.10	0.00
7.578	0.00	0.00	0.11	0.00
7.636	0.00	0.00	0.03	0.00
7.666	0.00	0.00	0.01	0.00
7.716	0.32	-0.01	0.00	0.02
7.772	0.00	0.00	-0.14	0.00
7.791	0.00	0.00	0.04	0.00
7.843	0.00	0.00	0.06	0.00
7.912	0.18	-0.10	0.00	0.06
7.966	0.00	0.00	0.03	0.00
7.974	0.12	0.01	0.00	-0.06
8.053	-0.31	-0.65	0.00	0.51
8.079	0.14	0.69	0.00	-0.41
8.111	-0.31	-0.20	0.00	-0.25
8.176	-0.01	0.07	0.00	0.00
8.239	0.00	0.00	0.02	0.00
8.261	-0.01	0.00	0.00	0.02
8.342	-0.41	-0.08	0.00	-0.20
8.387	0.00	0.00	-0.07	0.00
8.397	0.00	0.00	-0.10	0.00
8.411	0.73	0.08	0.00	0.30
8.424	0.00	0.00	0.08	0.00
8.434	-0.81	-0.37	0.00	-0.57
8.493	0.00	0.00	-0.19	0.00
8.565	0.43	0.78	0.00	0.79
8.565	0.00	0.00	0.16	0.00
8.588	-0.09	-0.21	0.00	-0.28
8.589	0.00	0.00	0.07	0.00
8.629	0.00	0.00	0.00	0.00
8.670	0.00	0.00	-0.02	0.00
8.751	-0.06	-0.02	0.00	0.05
8.828	-0.73	-0.67	0.00	-0.71
8.853	0.00	0.00	0.00	0.00
8.880	0.86	0.61	0.00	0.74
8.892	0.00	0.00	-0.06	0.00
8.904	-0.13	-0.12	0.00	0.17
8.937	0.66	1.64	0.00	-1.59
8.963	-0.51	-0.33	0.00	0.55
8.985	0.00	0.00	0.11	0.00
9.000	-0.20	-0.86	0.00	0.50

9.023	0.00	0.00	-0.09	0.00
9.099	0.00	-0.05	0.00	0.01
9.113	0.00	0.00	-0.03	0.00
9.150	0.00	0.00	0.01	0.00
9.163	-0.06	0.02	0.00	0.17
9.221	-0.01	0.00	0.00	-0.01
9.225	0.00	0.00	0.00	0.00
9.263	0.00	0.00	0.00	0.00
9.348	-0.12	0.17	0.00	-0.18
9.360	0.06	-0.37	0.00	0.26
9.400	-0.03	0.06	0.00	-0.02
9.409	0.00	0.00	-0.02	0.00
9.484	0.00	0.00	-0.02	0.00
9.488	0.02	-0.25	0.00	-0.01
9.515	0.00	0.00	0.04	0.00
9.555	0.01	0.07	0.00	-0.04
9.572	0.00	0.00	0.00	0.00
9.627	0.00	0.00	0.00	0.00
9.652	0.16	-0.05	0.00	0.02
9.680	0.71	-0.01	0.00	-0.02
9.684	-0.61	-0.02	0.00	0.12
9.774	-0.20	0.04	0.00	-0.06
9.777	0.00	0.00	-0.04	0.00
9.792	0.00	0.00	0.05	0.00
9.828	0.00	0.00	-0.25	0.00
9.836	0.00	0.00	0.15	0.00

Table S4. Contribution of the k[th] excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 2. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	-0.02	0.00
4.989	2.89	0.32	0.00	-1.60
5.253	-0.61	0.57	0.00	0.37
5.395	0.00	0.00	-0.06	0.00
5.555	0.00	0.00	-0.08	0.00
5.660	0.00	0.00	-0.04	0.00
5.852	0.00	0.00	0.03	0.00
6.169	-2.08	-14.10	0.00	5.54
6.206	0.06	8.62	0.00	-1.57
6.253	0.00	5.72	0.00	-2.67
6.254	0.00	0.00	0.00	0.00
6.506	0.00	0.00	0.00	0.00
6.560	0.00	0.00	0.49	0.00
6.572	0.43	0.91	0.00	0.75
6.595	0.00	0.00	0.70	0.00

6.611	0.00	0.00	-2.11	0.00
6.617	0.00	0.00	1.16	0.00
6.646	-1.03	-1.10	0.00	-1.17
6.869	0.00	0.00	-0.20	0.00
6.888	-0.31	-0.25	0.00	-0.33
6.959	0.00	0.00	0.11	0.00
7.014	0.22	0.26	0.00	-0.67
7.072	-1.70	0.48	0.00	2.14
7.133	0.00	0.00	0.05	0.00
7.179	-0.58	0.17	0.00	-0.20
7.312	3.17	2.83	0.00	-5.09
7.332	0.00	0.00	0.00	0.00
7.370	2.25	3.29	0.00	-5.19
7.398	23.13	5.89	0.00	-16.45
7.406	-25.67	-13.96	0.00	26.76
7.477	0.00	0.00	-0.01	0.00
7.509	0.49	0.51	0.00	-0.67
7.537	0.00	0.00	-0.14	0.00
7.564	0.00	0.00	-0.12	0.00
7.578	0.00	0.00	0.21	0.00
7.636	0.00	0.00	0.07	0.00
7.666	0.00	0.00	0.02	0.00
7.716	-0.24	-0.06	0.00	0.26
7.772	0.00	0.00	-0.26	0.00
7.791	0.00	0.00	0.05	0.00
7.843	0.00	0.00	0.11	0.00
7.912	0.39	-0.41	0.00	0.04
7.966	0.00	0.00	0.06	0.00
7.974	-0.42	-0.01	0.00	0.07
8.053	-0.10	-1.55	0.00	0.96
8.079	0.28	1.86	0.00	-0.89
8.111	-0.31	-0.29	0.00	-0.30
8.176	-0.03	0.15	0.00	0.00
8.239	0.00	0.00	0.02	0.00
8.261	0.00	0.08	0.00	-0.01
8.342	-0.41	-0.08	0.00	-0.20
8.387	0.00	0.00	0.07	0.00
8.397	0.00	0.00	-0.21	0.00
8.411	1.73	0.40	0.00	0.86
8.424	0.00	0.00	0.13	0.00
8.434	-1.94	-0.91	0.00	-1.38
8.493	0.00	0.00	0.08	0.00
8.565	1.42	2.94	0.00	2.92
8.565	0.00	0.00	-0.02	0.00
8.588	-0.74	-1.45	0.00	-2.20
8.589	0.00	0.00	-0.07	0.00
8.629	0.00	0.00	-0.05	0.00
8.670	0.00	0.00	-0.02	0.00

8.751	-0.13	-0.16	0.00	0.15
8.828	-1.26	-0.22	0.00	-0.81
8.853	0.00	0.00	0.00	0.00
8.880	1.31	-0.09	0.00	0.58
8.892	0.00	0.00	-0.08	0.00
8.904	0.13	-0.21	0.00	0.23
8.937	0.89	2.34	0.00	-2.14
8.963	-0.44	-0.29	0.00	0.49
8.985	0.00	0.00	0.24	0.00
9.000	-0.27	-1.28	0.00	0.71
9.023	0.00	0.00	-0.19	0.00
9.099	-0.21	-0.69	0.00	0.38
9.113	0.00	0.00	0.00	0.00
9.150	0.00	0.00	0.04	0.00
9.163	0.12	0.02	0.00	0.11
9.221	-0.01	-0.04	0.00	-0.02
9.225	0.00	0.00	0.00	0.00
9.263	0.00	0.00	0.00	0.00
9.348	-0.57	1.31	0.00	-0.75
9.360	0.47	-1.30	0.00	0.83
9.400	-0.01	0.05	0.00	0.00
9.409	0.00	0.00	0.02	0.00
9.484	0.00	0.00	0.02	0.00
9.488	0.09	-0.76	0.00	0.00
9.515	0.00	0.00	-0.04	0.00
9.555	0.04	0.13	0.00	-0.08
9.572	0.00	0.00	0.00	0.00
9.627	0.00	0.00	0.05	0.00
9.652	0.44	0.05	0.00	-0.15
9.680	3.81	0.13	0.00	-0.75
9.684	-4.06	-0.13	0.00	0.79
9.774	0.15	0.18	0.00	0.17
9.777	0.00	0.00	0.16	0.00
9.792	0.00	0.00	-0.11	0.00
9.828	0.00	0.00	0.97	0.00
9.836	0.00	0.00	-0.86	0.00

Table S5. Contribution of the kth excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 3. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	-0.02	0.00
4.989	1.24	0.32	0.00	-1.51
5.253	-0.91	0.90	0.00	0.59
5.395	0.00	0.00	0.06	0.00
5.555	0.00	0.00	-0.25	0.00

5.660	0.00	0.00	0.18	0.00
5.852	0.00	0.00	0.03	0.00
6.169	-0.15	-22.32	0.00	5.49
6.206	-0.31	-5.93	0.00	7.87
6.253	0.25	27.94	0.00	-13.18
6.254	0.00	0.00	0.00	0.00
6.506	0.00	0.00	-0.04	0.00
6.560	0.00	0.00	1.02	0.00
6.572	-0.71	-1.51	0.00	-1.24
6.595	0.00	0.00	-1.01	0.00
6.611	0.00	0.00	-2.22	0.00
6.617	0.00	0.00	2.48	0.00
6.646	-1.49	-1.28	0.00	-1.59
6.869	0.00	0.00	-0.04	0.00
6.888	0.44	0.73	0.00	0.83
6.959	0.00	0.00	0.00	0.00
7.014	-1.06	0.31	0.00	3.06
7.072	1.91	-0.21	0.00	-0.85
7.133	0.00	0.00	0.00	0.00
7.179	-0.52	0.06	0.00	-0.23
7.312	-2.60	-7.67	0.00	5.10
7.332	0.00	0.00	0.00	0.00
7.370	0.38	5.83	0.00	-1.49
7.398	-41.41	-16.27	0.00	31.81
7.406	42.33	17.31	0.00	-34.79
7.477	0.00	0.00	0.01	0.00
7.509	0.22	0.56	0.00	-0.38
7.537	0.00	0.00	0.06	0.00
7.564	0.00	0.00	0.12	0.00
7.578	0.00	0.00	-0.18	0.00
7.636	0.00	0.00	0.06	0.00
7.666	0.00	0.00	0.04	0.00
7.716	2.51	0.15	0.00	-0.76
7.772	0.00	0.00	-0.43	0.00
7.791	0.00	0.00	0.14	0.00
7.843	0.00	0.00	0.17	0.00
7.912	0.09	-0.31	0.00	-0.08
7.966	0.00	0.00	0.06	0.00
7.974	0.18	-0.01	0.00	0.05
8.053	0.20	-0.69	0.00	0.32
8.079	0.15	0.59	0.00	-0.45
8.111	-0.51	-1.11	0.00	-0.88
8.176	-0.01	0.27	0.00	0.05
8.239	0.00	0.00	-0.05	0.00
8.261	-0.07	-0.08	0.00	0.17
8.342	1.94	1.22	0.00	1.54
8.387	0.00	0.00	0.14	0.00
8.397	0.00	0.00	-0.03	0.00

8.411	-0.30	-0.27	0.00	-0.31
8.424	0.00	0.00	-0.06	0.00
8.434	-1.35	-0.79	0.00	-1.12
8.493	0.00	0.00	-0.41	0.00
8.565	-0.99	-3.55	0.00	-3.32
8.565	0.00	0.00	0.37	0.00
8.588	1.01	4.13	0.00	3.16
8.589	0.00	0.00	0.02	0.00
8.629	0.00	0.00	-0.04	0.00
8.670	0.00	0.00	0.02	0.00
8.751	0.00	0.16	0.00	-0.06
8.828	-1.46	-0.52	0.00	-1.06
8.853	0.00	0.00	0.00	0.00
8.880	1.89	0.28	0.00	1.05
8.892	0.00	0.00	-0.08	0.00
8.904	-1.64	-0.73	0.00	1.22
8.937	-0.50	-1.96	0.00	1.28
8.963	0.27	-0.28	0.00	0.34
8.985	0.00	0.00	0.19	0.00
9.000	0.42	1.71	0.00	-1.07
9.023	0.00	0.00	-0.21	0.00
9.099	0.51	1.87	0.00	-0.98
9.113	0.00	0.00	0.00	0.00
9.150	0.00	0.00	0.04	0.00
9.163	0.47	0.00	0.00	0.00
9.221	0.02	-0.11	0.00	0.01
9.225	0.00	0.00	0.00	0.00
9.263	0.00	0.00	0.00	0.00
9.348	-0.02	0.30	0.00	0.01
9.360	0.15	-0.17	0.00	0.08
9.400	0.00	0.06	0.00	0.01
9.409	0.00	0.00	-0.05	0.00
9.484	0.00	0.00	-0.08	0.00
9.488	-0.02	-0.69	0.00	-0.15
9.515	0.00	0.00	0.08	0.00
9.555	0.06	0.15	0.00	-0.09
9.572	0.00	0.00	0.00	0.00
9.627	0.00	0.00	0.05	0.00
9.652	1.07	0.07	0.00	-0.32
9.680	-11.60	-0.48	0.00	2.54
9.684	10.81	0.38	0.00	-2.14
9.774	-0.19	0.04	0.00	-0.06
9.777	0.00	0.00	-0.28	0.00
9.792	0.00	0.00	0.40	0.00
9.828	0.00	0.00	1.22	0.00
9.836	0.00	0.00	-0.96	0.00

Table S6. Contribution of the k[th] excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 4. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	0.02	0.00
4.989	-2.89	0.50	0.00	-2.11
5.253	0.91	0.90	0.00	1.08
5.395	0.00	0.00	-0.19	0.00
5.555	0.00	0.00	0.03	0.00
5.660	0.00	0.00	0.18	0.00
5.852	0.00	0.00	-0.08	0.00
6.169	-2.08	-20.30	0.00	7.02
6.206	0.34	35.29	0.00	-8.97
6.253	1.24	-11.31	0.00	4.62
6.254	0.00	0.00	-0.02	0.00
6.506	0.00	0.00	-0.02	0.00
6.560	0.00	0.00	-0.97	0.00
6.572	0.71	0.24	0.00	0.43
6.595	0.00	0.00	-0.39	0.00
6.611	0.00	0.00	1.78	0.00
6.617	0.00	0.00	-0.66	0.00
6.646	4.02	-6.95	0.00	1.02
6.869	0.00	0.00	0.44	0.00
6.888	2.18	-0.06	0.00	0.49
6.959	0.00	0.00	-0.05	0.00
7.014	1.46	-1.03	0.00	-4.15
7.072	-5.52	0.30	0.00	1.08
7.133	0.00	0.00	0.00	0.00
7.179	-0.35	-1.32	0.00	-0.84
7.312	-0.37	8.85	0.00	-0.99
7.332	0.00	0.00	0.00	0.00
7.370	-4.94	-20.35	0.00	12.93
7.398	79.75	39.05	0.00	-64.45
7.406	-73.69	-25.16	0.00	52.42
7.477	0.00	0.00	0.02	0.00
7.509	-0.04	0.05	0.00	0.04
7.537	0.00	0.00	0.19	0.00
7.564	0.00	0.00	0.24	0.00
7.578	0.00	0.00	-0.35	0.00
7.636	0.00	0.00	-0.03	0.00
7.666	0.00	0.00	0.04	0.00
7.716	-0.24	-0.05	0.00	0.22
7.772	0.00	0.00	-0.12	0.00
7.791	0.00	0.00	0.03	0.00
7.843	0.00	0.00	0.07	0.00
7.912	-0.98	0.21	0.00	-0.48

7.966	0.00	0.00	-0.09	0.00
7.974	1.14	-0.07	0.00	0.60
8.053	-1.53	3.41	0.00	-1.37
8.079	-0.50	-3.18	0.00	1.58
8.111	0.75	-0.63	0.00	-0.06
8.176	0.46	0.92	0.00	0.73
8.239	0.00	0.00	-0.11	0.00
8.261	0.22	-0.72	0.00	-0.46
8.342	-1.53	0.23	0.00	-0.42
8.387	0.00	0.00	-0.79	0.00
8.397	0.00	0.00	0.53	0.00
8.411	-1.30	-0.02	0.00	-0.43
8.424	0.00	0.00	0.17	0.00
8.434	3.11	1.65	0.00	2.40
8.493	0.00	0.00	-0.41	0.00
8.565	0.52	1.94	0.00	1.81
8.565	0.00	0.00	0.35	0.00
8.588	-0.60	-2.69	0.00	-1.89
8.589	0.00	0.00	0.36	0.00
8.629	0.00	0.00	-0.06	0.00
8.670	0.00	0.00	0.04	0.00
8.751	0.16	-0.49	0.00	0.05
8.828	2.98	-0.60	0.00	1.42
8.853	0.00	0.00	-0.01	0.00
8.880	-5.00	-0.28	0.00	-2.54
8.892	0.00	0.00	-0.06	0.00
8.904	2.11	2.87	0.00	-3.93
8.937	0.30	0.51	0.00	-0.69
8.963	-0.31	-2.18	0.00	3.11
8.985	0.00	0.00	0.21	0.00
9.000	-0.29	-1.59	0.00	0.78
9.023	0.00	0.00	-0.17	0.00
9.099	0.74	-0.30	0.00	-0.72
9.113	0.00	0.00	-0.05	0.00
9.150	0.00	0.00	0.07	0.00
9.163	-1.12	0.05	0.00	0.33
9.221	0.04	0.39	0.00	0.12
9.225	0.00	0.00	0.02	0.00
9.263	0.00	0.00	0.00	0.00
9.348	1.22	-2.97	0.00	1.57
9.360	-1.26	2.89	0.00	-1.77
9.400	-0.04	-0.02	0.00	-0.05
9.409	0.00	0.00	0.02	0.00
9.484	0.00	0.00	0.25	0.00
9.488	-0.07	0.69	0.00	0.03
9.515	0.00	0.00	-0.22	0.00
9.555	-0.01	-0.32	0.00	0.13
9.572	0.00	0.00	0.00	0.00

9.627	0.00	0.00	-0.14	0.00
9.652	-2.91	-0.10	0.00	0.75
9.680	23.86	0.96	0.00	-5.15
9.684	-22.51	-0.86	0.00	4.58
9.774	-0.25	-0.07	0.00	-0.15
9.777	0.00	0.00	0.16	0.00
9.792	0.00	0.00	1.11	0.00
9.828	0.00	0.00	-2.75	0.00
9.836	0.00	0.00	1.92	0.00

Table S7. Contribution of the k[th] excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 5. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	-0.02	0.00
4.989	2.07	0.27	0.00	-1.35
5.253	-1.06	-0.41	0.00	-0.67
5.395	0.00	0.00	-0.06	0.00
5.555	0.00	0.00	-0.08	0.00
5.660	0.00	0.00	0.09	0.00
5.852	0.00	0.00	-0.05	0.00
6.169	-0.96	-18.29	0.00	5.38
6.206	-0.06	6.73	0.00	1.41
6.253	0.25	9.31	0.00	-4.48
6.254	0.00	0.00	0.00	0.00
6.506	0.00	0.00	-0.04	0.00
6.560	0.00	0.00	0.34	0.00
6.572	-0.47	-0.65	0.00	-0.60
6.595	0.00	0.00	-0.16	0.00
6.611	0.00	0.00	-0.44	0.00
6.617	0.00	0.00	0.25	0.00
6.646	-0.11	2.74	0.00	0.77
6.869	0.00	0.00	0.06	0.00
6.888	0.06	0.10	0.00	0.11
6.959	0.00	0.00	0.08	0.00
7.014	-0.46	0.36	0.00	1.31
7.072	0.00	-0.44	0.00	-2.02
7.133	0.00	0.00	0.00	0.00
7.179	-0.35	-0.17	0.00	-0.26
7.312	-1.99	-2.24	0.00	3.28
7.332	0.00	0.00	-0.01	0.00
7.370	-0.54	-0.19	0.00	1.17
7.398	-33.68	-11.70	0.00	25.24
7.406	35.08	14.26	0.00	-28.68
7.477	0.00	0.00	-0.01	0.00
7.509	0.00	-0.10	0.00	0.02

7.537	0.00	0.00	0.05	0.00
7.564	0.00	0.00	0.13	0.00
7.578	0.00	0.00	-0.19	0.00
7.636	0.00	0.00	-0.03	0.00
7.666	0.00	0.00	-0.04	0.00
7.716	0.73	0.05	0.00	-0.25
7.772	0.00	0.00	-0.09	0.00
7.791	0.00	0.00	0.04	0.00
7.843	0.00	0.00	0.07	0.00
7.912	0.33	0.00	0.00	0.19
7.966	0.00	0.00	-0.03	0.00
7.974	-0.18	0.02	0.00	-0.15
8.053	0.00	-0.22	0.00	0.13
8.079	0.05	0.46	0.00	-0.18
8.111	0.03	-0.11	0.00	-0.05
8.176	-0.05	0.02	0.00	-0.06
8.239	0.00	0.00	0.02	0.00
8.261	-0.07	0.00	0.00	0.16
8.342	1.33	0.38	0.00	0.75
8.387	0.00	0.00	0.17	0.00
8.397	0.00	0.00	-0.18	0.00
8.411	-0.53	-0.08	0.00	-0.23
8.424	0.00	0.00	0.01	0.00
8.434	-0.09	0.00	0.00	-0.02
8.493	0.00	0.00	0.19	0.00
8.565	-0.24	-0.39	0.00	-0.40
8.565	0.00	0.00	-0.14	0.00
8.588	0.00	0.41	0.00	0.04
8.589	0.00	0.00	-0.04	0.00
8.629	0.00	0.00	-0.05	0.00
8.670	0.00	0.00	0.02	0.00
8.751	0.00	0.05	0.00	-0.02
8.828	-0.13	0.52	0.00	0.16
8.853	0.00	0.00	0.00	0.00
8.880	0.06	-0.28	0.00	-0.11
8.892	0.00	0.00	0.00	0.00
8.904	-0.55	-0.47	0.00	0.68
8.937	-0.32	-1.07	0.00	0.79
8.963	0.14	0.02	0.00	-0.06
8.985	0.00	0.00	0.12	0.00
9.000	0.29	0.67	0.00	-0.66
9.023	0.00	0.00	-0.05	0.00
9.099	0.14	0.49	0.00	-0.26
9.113	0.00	0.00	0.00	0.00
9.150	0.00	0.00	0.04	0.00
9.163	0.19	-0.02	0.00	-0.10
9.221	0.00	-0.01	0.00	0.00
9.225	0.00	0.00	0.00	0.00

9.263	0.00	0.00	0.00	0.00
9.348	-0.12	-0.03	0.00	-0.21
9.360	0.13	-0.23	0.00	0.13
9.400	0.00	0.03	0.00	0.00
9.409	0.00	0.00	-0.05	0.00
9.484	0.00	0.00	0.02	0.00
9.488	-0.04	0.38	0.00	0.00
9.515	0.00	0.00	0.00	0.00
9.555	0.03	-0.02	0.00	-0.01
9.572	0.00	0.00	0.00	0.00
9.627	0.00	0.00	0.00	0.00
9.652	0.85	0.06	0.00	-0.26
9.680	-8.82	-0.34	0.00	1.83
9.684	8.19	0.28	0.00	-1.61
9.774	-0.13	-0.04	0.00	-0.08
9.777	0.00	0.00	0.20	0.00
9.792	0.00	0.00	-0.11	0.00
9.828	0.00	0.00	0.99	0.00
9.836	0.00	0.00	-1.11	0.00

Table S8. Contribution of the k[th] excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 6. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	-0.02	0.00
4.989	-1.24	0.05	0.00	-0.14
5.253	0.76	-1.55	0.00	-1.24
5.395	0.00	0.00	0.32	0.00
5.555	0.00	0.00	0.11	0.00
5.660	0.00	0.00	-0.27	0.00
5.852	0.00	0.00	0.00	0.00
6.169	0.74	18.60	0.00	-5.22
6.206	0.22	11.04	0.00	-5.63
6.253	-0.99	-26.08	0.00	12.71
6.254	0.00	0.00	0.02	0.00
6.506	0.00	0.00	-0.04	0.00
6.560	0.00	0.00	0.54	0.00
6.572	0.43	0.73	0.00	0.63
6.595	0.00	0.00	0.08	0.00
6.611	0.00	0.00	0.90	0.00
6.617	0.00	0.00	-1.57	0.00
6.646	0.80	-3.11	0.00	-0.34
6.869	0.00	0.00	0.04	0.00
6.888	0.31	-0.16	0.00	-0.08
6.959	0.00	0.00	0.05	0.00
7.014	0.14	0.67	0.00	-0.47

7.072	3.82	-0.67	0.00	-2.87
7.133	0.00	0.00	-0.03	0.00
7.179	-0.81	0.17	0.00	-0.31
7.312	-4.10	-3.78	0.00	6.61
7.332	0.00	0.00	0.01	0.00
7.370	-3.66	-9.87	0.00	8.97
7.398	32.91	20.22	0.00	-28.29
7.406	-27.05	-6.11	0.00	14.13
7.477	0.00	0.00	-0.01	0.00
7.509	-0.09	-0.41	0.00	0.19
7.537	0.00	0.00	0.03	0.00
7.564	0.00	0.00	0.05	0.00
7.578	0.00	0.00	-0.14	0.00
7.636	0.00	0.00	-0.01	0.00
7.666	0.00	0.00	0.04	0.00
7.716	-0.57	-0.01	0.00	0.08
7.772	0.00	0.00	0.38	0.00
7.791	0.00	0.00	-0.14	0.00
7.843	0.00	0.00	-0.21	0.00
7.912	-0.45	0.17	0.00	-0.18
7.966	0.00	0.00	0.00	0.00
7.974	-0.12	-0.05	0.00	0.43
8.053	0.51	1.68	0.00	-1.21
8.079	-0.31	-1.74	0.00	0.95
8.111	0.58	0.46	0.00	0.51
8.176	0.14	-0.15	0.00	0.13
8.239	0.00	0.00	0.02	0.00
8.261	0.08	0.08	0.00	-0.21
8.342	-1.74	-0.84	0.00	-1.21
8.387	0.00	0.00	-0.41	0.00
8.397	0.00	0.00	0.31	0.00
8.411	-0.07	0.17	0.00	0.11
8.424	0.00	0.00	0.17	0.00
8.434	1.35	0.47	0.00	0.80
8.493	0.00	0.00	-0.04	0.00
8.565	0.24	-0.25	0.00	-0.14
8.565	0.00	0.00	-0.06	0.00
8.588	0.20	1.14	0.00	0.67
8.589	0.00	0.00	0.04	0.00
8.629	0.00	0.00	0.00	0.00
8.670	0.00	0.00	0.02	0.00
8.751	0.00	-0.14	0.00	0.05
8.828	-0.40	-2.17	0.00	-1.18
8.853	0.00	0.00	0.00	0.00
8.880	0.26	0.65	0.00	0.46
8.892	0.00	0.00	0.03	0.00
8.904	-0.55	0.17	0.00	-0.10
8.937	-0.39	0.19	0.00	0.80

8.963	0.24	0.43	0.00	-0.64
8.985	0.00	0.00	-0.29	0.00
9.000	-0.02	-0.37	0.00	0.08
9.023	0.00	0.00	0.28	0.00
9.099	-0.16	-0.10	0.00	0.20
9.113	0.00	0.00	0.05	0.00
9.150	0.00	0.00	0.01	0.00
9.163	-0.34	0.01	0.00	0.03
9.221	0.01	0.08	0.00	0.03
9.225	0.00	0.00	0.01	0.00
9.263	0.00	0.00	0.00	0.00
9.348	0.58	-1.21	0.00	0.78
9.360	-0.60	1.35	0.00	-0.83
9.400	0.08	-0.01	0.00	0.10
9.409	0.00	0.00	0.00	0.00
9.484	0.00	0.00	0.05	0.00
9.488	0.15	-0.32	0.00	0.17
9.515	0.00	0.00	-0.08	0.00
9.555	0.03	-0.07	0.00	0.01
9.572	0.00	0.00	0.00	0.00
9.627	0.00	0.00	-0.01	0.00
9.652	0.11	0.04	0.00	-0.07
9.680	10.78	0.49	0.00	-2.51
9.684	-11.32	-0.39	0.00	2.23
9.774	0.55	0.09	0.00	0.30
9.777	0.00	0.00	-0.08	0.00
9.792	0.00	0.00	0.27	0.00
9.828	0.00	0.00	-0.21	0.00
9.836	0.00	0.00	0.08	0.00

Table S9. Contribution of the k[th] excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 7. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	0.02	0.00
4.989	0.00	-0.27	0.00	1.24
5.253	-3.64	0.08	0.00	-0.91
5.395	0.00	0.00	-0.06	0.00
5.555	0.00	0.00	-0.03	0.00
5.660	0.00	0.00	-0.04	0.00
5.852	0.00	0.00	0.02	0.00
6.169	-0.22	-22.32	0.00	5.57
6.206	0.40	25.59	0.00	-10.22
6.253	0.75	-9.58	0.00	4.07
6.254	0.00	0.00	0.02	0.00
6.506	0.00	0.00	0.02	0.00

6.560	0.00	0.00	-0.68	0.00
6.572	0.05	0.73	0.00	0.48
6.595	0.00	0.00	1.09	0.00
6.611	0.00	0.00	0.53	0.00
6.617	0.00	0.00	-0.91	0.00
6.646	-0.69	2.01	0.00	0.08
6.869	0.00	0.00	0.04	0.00
6.888	-0.44	-0.48	0.00	-0.58
6.959	0.00	0.00	0.05	0.00
7.014	-0.08	0.62	0.00	0.18
7.072	4.04	0.02	0.00	0.32
7.133	0.00	0.00	0.00	0.00
7.179	-0.47	-0.52	0.00	-0.49
7.312	3.57	8.26	0.00	-6.61
7.332	0.00	0.00	-0.05	0.00
7.370	1.29	-2.11	0.00	-2.51
7.398	26.39	9.84	0.00	-20.06
7.406	-33.71	-15.34	0.00	30.24
7.477	0.00	0.00	-0.02	0.00
7.509	0.62	-0.46	0.00	-0.61
7.537	0.00	0.00	-0.09	0.00
7.564	0.00	0.00	-0.05	0.00
7.578	0.00	0.00	0.06	0.00
7.636	0.00	0.00	-0.01	0.00
7.666	0.00	0.00	-0.07	0.00
7.716	-2.27	-0.14	0.00	0.71
7.772	0.00	0.00	0.14	0.00
7.791	0.00	0.00	-0.08	0.00
7.843	0.00	0.00	0.03	0.00
7.912	0.80	-0.07	0.00	0.43
7.966	0.00	0.00	0.00	0.00
7.974	-0.90	0.05	0.00	-0.40
8.053	-0.31	-1.55	0.00	1.05
8.079	0.18	1.71	0.00	-0.61
8.111	0.31	1.28	0.00	0.89
8.176	0.00	-0.12	0.00	-0.03
8.239	0.00	0.00	0.11	0.00
8.261	-0.09	0.12	0.00	0.21
8.342	5.00	1.60	0.00	2.94
8.387	0.00	0.00	0.07	0.00
8.397	0.00	0.00	-0.13	0.00
8.411	-0.17	-0.15	0.00	-0.17
8.424	0.00	0.00	0.18	0.00
8.434	-2.57	-1.00	0.00	-1.63
8.493	0.00	0.00	0.41	0.00
8.565	-0.52	0.69	0.00	0.44
8.565	0.00	0.00	-0.39	0.00
8.588	-0.37	-1.65	0.00	-1.18

8.589	0.00	0.00	-0.31	0.00
8.629	0.00	0.00	0.08	0.00
8.670	0.00	0.00	-0.07	0.00
8.751	0.00	0.61	0.00	-0.21
8.828	0.86	2.47	0.00	1.58
8.853	0.00	0.00	0.00	0.00
8.880	0.16	-1.97	0.00	-0.95
8.892	0.00	0.00	0.11	0.00
8.904	0.46	-1.68	0.00	1.95
8.937	-0.24	-0.82	0.00	0.60
8.963	-0.07	1.68	0.00	-2.34
8.985	0.00	0.00	-0.08	0.00
9.000	0.26	1.34	0.00	-0.69
9.023	0.00	0.00	0.05	0.00
9.099	-0.23	0.30	0.00	0.18
9.113	0.00	0.00	0.08	0.00
9.150	0.00	0.00	-0.07	0.00
9.163	0.50	-0.04	0.00	-0.26
9.221	0.02	-0.12	0.00	0.01
9.225	0.00	0.00	-0.02	0.00
9.263	0.00	0.00	-0.01	0.00
9.348	-0.78	2.26	0.00	-0.96
9.360	0.81	-1.85	0.00	1.13
9.400	0.01	-0.01	0.00	0.01
9.409	0.00	0.00	0.05	0.00
9.484	0.00	0.00	-0.16	0.00
9.488	-0.07	-0.51	0.00	-0.18
9.515	0.00	0.00	0.18	0.00
9.555	-0.14	0.48	0.00	-0.10
9.572	0.00	0.00	0.01	0.00
9.627	0.00	0.00	0.02	0.00
9.652	0.93	-0.04	0.00	-0.16
9.680	-22.22	-0.81	0.00	4.49
9.684	21.91	0.79	0.00	-4.37
9.774	0.28	0.09	0.00	0.17
9.777	0.00	0.00	0.68	0.00
9.792	0.00	0.00	-1.43	0.00
9.828	0.00	0.00	0.64	0.00
9.836	0.00	0.00	0.06	0.00

Table S10. Contribution of the kth excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 8. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	0.02	0.00
4.989	-19.82	1.04	0.00	-3.66

5.253	10.32	-1.47	0.00	1.41
5.395	0.00	0.00	0.00	0.00
5.395	0.00	0.00	0.30	0.00
5.395	0.00	0.00	-0.27	0.00
5.395	0.00	0.00	-0.08	0.00
6.169	-0.67	12.55	0.00	-2.30
6.206	-0.29	-22.90	0.00	7.64
6.253	-0.25	1.73	0.00	-0.67
6.254	0.00	0.00	-0.04	0.00
6.506	0.00	0.00	0.11	0.00
6.560	0.00	0.00	0.68	0.00
6.572	-0.99	-0.19	0.00	-0.51
6.595	0.00	0.00	-1.52	0.00
6.611	0.00	0.00	4.68	0.00
6.617	0.00	0.00	-3.92	0.00
6.646	3.79	6.22	0.00	4.97
6.869	0.00	0.00	0.36	0.00
6.888	1.06	-0.25	0.00	0.02
6.959	0.00	0.00	-0.05	0.00
7.014	0.88	-0.15	0.00	-2.56
7.072	1.49	-0.18	0.00	-0.77
7.133	0.00	0.00	-0.03	0.00
7.179	0.93	0.40	0.00	0.66
7.312	6.37	14.16	0.00	-11.70
7.332	0.00	0.00	-0.06	0.00
7.370	1.09	-7.38	0.00	-1.46
7.398	81.73	31.54	0.00	-62.55
7.406	-91.33	-34.81	0.00	70.89
7.477	0.00	0.00	0.06	0.00
7.509	-0.80	-0.31	0.00	0.98
7.537	0.00	0.00	-0.22	0.00
7.564	0.00	0.00	-0.06	0.00
7.578	0.00	0.00	0.22	0.00
7.636	0.00	0.00	-0.09	0.00
7.666	0.00	0.00	-0.14	0.00
7.716	-5.43	-0.29	0.00	1.51
7.772	0.00	0.00	0.96	0.00
7.791	0.00	0.00	-0.27	0.00
7.843	0.00	0.00	-0.37	0.00
7.912	-0.89	0.62	0.00	-0.25
7.966	0.00	0.00	-0.21	0.00
7.974	0.30	-0.02	0.00	0.14
8.053	-0.66	1.81	0.00	-0.79
8.079	-0.48	-2.13	0.00	1.43
8.111	1.02	1.43	0.00	1.28
8.176	0.17	-0.56	0.00	0.07
8.239	0.00	0.00	0.06	0.00
8.261	-0.02	0.64	0.00	-0.03

8.342	7.25	2.21	0.00	4.18
8.387	0.00	0.00	-0.24	0.00
8.397	0.00	0.00	0.42	0.00
8.411	-3.76	-1.28	0.00	-2.20
8.424	0.00	0.00	-0.06	0.00
8.434	1.35	0.70	0.00	1.03
8.493	0.00	0.00	0.87	0.00
8.565	-3.31	-3.80	0.00	-4.21
8.565	0.00	0.00	-0.72	0.00
8.588	0.34	-0.21	0.00	0.91
8.589	0.00	0.00	-0.09	0.00
8.629	0.00	0.00	0.06	0.00
8.670	0.00	0.00	-0.12	0.00
8.751	0.06	-0.49	0.00	0.12
8.828	3.97	5.40	0.00	4.63
8.853	0.00	0.00	0.00	0.00
8.880	-5.48	-2.34	0.00	-3.85
8.892	0.00	0.00	0.17	0.00
8.904	4.55	-0.31	0.00	-0.56
8.937	-0.14	-1.39	0.00	0.45
8.963	1.40	1.84	0.00	-2.83
8.985	0.00	0.00	-0.30	0.00
9.000	-0.14	-1.16	0.00	0.42
9.023	0.00	0.00	0.14	0.00
9.099	-0.42	-1.67	0.00	0.84
9.113	0.00	0.00	0.06	0.00
9.150	0.00	0.00	-0.14	0.00
9.163	0.22	-0.06	0.00	-0.41
9.221	0.03	0.01	0.00	0.04
9.225	0.00	0.00	0.00	0.00
9.263	0.00	0.00	0.00	0.00
9.348	0.48	-1.21	0.00	0.61
9.360	-0.32	0.99	0.00	-0.64
9.400	0.02	-0.03	0.00	0.02
9.409	0.00	0.00	-0.02	0.00
9.484	0.00	0.00	-0.05	0.00
9.488	0.02	0.51	0.00	0.12
9.515	0.00	0.00	0.06	0.00
9.555	-0.01	-0.01	0.00	0.01
9.572	0.00	0.00	0.00	0.00
9.627	0.00	0.00	-0.04	0.00
9.652	0.11	0.18	0.00	-0.23
9.680	-15.31	-0.68	0.00	3.53
9.684	15.25	0.45	0.00	-2.85
9.774	-0.23	-0.07	0.00	-0.14
9.777	0.00	0.00	0.16	0.00
9.792	0.00	0.00	-0.98	0.00
9.828	0.00	0.00	-1.65	0.00

9.836	0.00	0.00	1.99	0.00

Table S11. Contribution of the k[th] excited electronic state in the sum over state summation of the normal mode derivative of the polarizability for the normal mode vibration assigned to peak 9. The $\frac{\partial \alpha_{\gamma\sigma}(\lambda_{ex})}{\partial Q_j}$ are in atomic units and λ_{ex}=785 nm.

$E_k - E_0$ (eV)	$\frac{\partial \alpha_{XX}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{YY}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{ZZ}(\lambda_{ex})}{\partial Q_j}$	$\frac{\partial \alpha_{XY}(\lambda_{ex})}{\partial Q_j}$
4.930	0.00	0.00	0.06	0.00
4.989	-14.04	-0.82	0.00	4.49
5.253	6.98	-0.90	0.00	1.05
5.395	0.00	0.00	0.19	0.00
5.555	0.00	0.00	0.25	0.00
5.660	0.00	0.00	-0.18	0.00
5.852	0.00	0.00	-0.02	0.00
6.169	3.63	56.42	0.00	-17.29
6.206	-0.20	-40.94	0.00	5.53
6.253	-3.98	-18.36	0.00	10.70
6.254	0.00	0.00	-0.08	0.00
6.506	0.00	0.00	0.05	0.00
6.560	0.00	0.00	-2.00	0.00
6.572	0.19	0.13	0.00	0.16
6.595	0.00	0.00	0.08	0.00
6.611	0.00	0.00	8.52	0.00
6.617	0.00	0.00	-6.18	0.00
6.646	4.70	1.10	0.00	4.09
6.869	0.00	0.00	0.08	0.00
6.888	-0.12	-0.73	0.00	-0.75
6.959	0.00	0.00	-0.05	0.00
7.014	0.74	0.05	0.00	-2.16
7.072	2.76	-0.16	0.00	-0.59
7.133	0.00	0.00	0.00	0.00
7.179	0.99	0.40	0.00	0.69
7.312	-0.53	2.95	0.00	0.26
7.332	0.00	0.00	0.05	0.00
7.370	-2.16	-8.13	0.00	5.57
7.398	24.98	13.02	0.00	-20.52
7.406	-24.69	-6.11	0.00	13.77
7.477	0.00	0.00	0.03	0.00
7.509	0.33	-0.41	0.00	-0.29
7.537	0.00	0.00	-0.14	0.00
7.564	0.00	0.00	-0.33	0.00
7.578	0.00	0.00	0.49	0.00
7.636	0.00	0.00	-0.04	0.00
7.666	0.00	0.00	-0.01	0.00
7.716	-2.68	-0.10	0.00	0.57
7.772	0.00	0.00	1.04	0.00

7.791	0.00	0.00	-0.32	0.00
7.843	0.00	0.00	-0.32	0.00
7.912	0.27	-0.24	0.00	0.05
7.966	0.00	0.00	-0.49	0.00
7.974	-0.48	0.04	0.00	-0.29
8.053	1.58	0.00	0.00	-0.67
8.079	-0.17	-1.09	0.00	0.53
8.111	-1.16	1.85	0.00	0.62
8.176	-0.09	-0.49	0.00	-0.20
8.239	0.00	0.00	-0.08	0.00
8.261	0.13	0.20	0.00	-0.34
8.342	-0.51	-1.14	0.00	-0.96
8.387	0.00	0.00	-0.38	0.00
8.397	0.00	0.00	0.39	0.00
8.411	1.16	0.31	0.00	0.62
8.424	0.00	0.00	-0.05	0.00
8.434	-2.07	-0.96	0.00	-1.46
8.493	0.00	0.00	0.19	0.00
8.565	-0.43	-1.44	0.00	-1.35
8.565	0.00	0.00	-0.47	0.00
8.588	0.35	1.65	0.00	1.13
8.589	0.00	0.00	-0.07	0.00
8.629	0.00	0.00	0.08	0.00
8.670	0.00	0.00	0.06	0.00
8.751	0.00	-0.16	0.00	0.06
8.828	0.60	-1.42	0.00	-0.29
8.853	0.00	0.00	-0.01	0.00
8.880	0.45	1.41	0.00	0.95
8.892	0.00	0.00	0.06	0.00
8.904	0.25	-0.29	0.00	0.31
8.937	0.15	1.39	0.00	-0.48
8.963	-0.10	1.76	0.00	-2.46
8.985	0.00	0.00	-0.41	0.00
9.000	-0.29	-0.98	0.00	0.70
9.023	0.00	0.00	0.43	0.00
9.099	-0.77	-1.57	0.00	1.18
9.113	0.00	0.00	-0.02	0.00
9.150	0.00	0.00	0.04	0.00
9.163	-0.25	0.03	0.00	0.18
9.221	0.02	0.04	0.00	0.04
9.225	0.00	0.00	0.01	0.00
9.263	0.00	0.00	0.00	0.00
9.348	0.53	-1.21	0.00	0.69
9.360	-0.66	1.42	0.00	-0.86
9.400	0.06	0.11	0.00	0.10
9.409	0.00	0.00	0.07	0.00
9.484	0.00	0.00	0.02	0.00
9.488	0.11	-0.88	0.00	0.00

9.515	0.00	0.00	-0.06	0.00
9.555	-0.01	0.18	0.00	-0.06
9.572	0.00	0.00	-0.01	0.00
9.627	0.00	0.00	0.07	0.00
9.652	0.60	0.01	0.00	-0.15
9.680	-6.15	-0.16	0.00	1.02
9.684	5.49	0.19	0.00	-1.08
9.774	0.15	0.02	0.00	0.08
9.777	0.00	0.00	0.08	0.00
9.792	0.00	0.00	-0.56	0.00
9.828	0.00	0.00	-0.85	0.00
9.836	0.00	0.00	0.88	0.00

The XYZ of the calculated adenine clusters

XYZ geometries in Å are given for the adenine clusters calculated by B3LYP-D2/6-311+G(d,p).

adenine	X	Y	Z
N	-1.949	0.521	0.000
C	-1.299	1.696	0.000
N	0.017	1.920	0.000
C	0.707	0.776	0.000
C	0.179	-0.517	0.000
C	-1.226	-0.610	0.000
N	1.181	-1.470	0.000
C	2.287	-0.772	0.000
N	2.071	0.591	0.000
N	-1.864	-1.802	0.000
H	-1.933	2.578	0.000
H	3.286	-1.184	0.000
H	2.766	1.322	0.000
H	-2.870	-1.822	0.000
H	-1.333	-2.657	0.000

adenine H-bond dimer	X	Y	Z
N	-3.678	-1.945	-0.152
C	-4.874	-1.347	-0.106
N	-5.151	-0.044	-0.005
C	-4.033	0.683	0.047
C	-2.718	0.213	0.005
C	-2.561	-1.190	-0.094
N	-1.823	1.270	0.075
C	-2.573	2.340	0.159
N	-3.916	2.052	0.148
N	-1.367	-1.801	-0.137
H	-5.731	-2.013	-0.158
H	-2.209	3.355	0.230
H	-4.683	2.705	0.199
H	-1.375	-2.807	-0.197

H	-0.471	-1.309	-0.045
N	1.114	1.470	-0.144
C	1.924	0.408	-0.032
N	1.397	-0.828	0.105
C	2.224	-1.881	0.217
N	3.555	-1.899	0.211
C	4.056	-0.664	0.071
C	3.332	0.521	-0.055
N	4.171	1.612	-0.179
C	5.374	1.100	-0.132
N	5.373	-0.273	0.019
H	1.735	-2.845	0.327
H	6.295	1.659	-0.200
H	6.173	-0.883	0.081
H	0.095	1.368	-0.072
H	1.536	2.380	-0.230

Adenine π-stacked dimer	X	Y	Z
H	2.782	-2.906	0.136
C	2.367	-1.942	-0.145
H	-0.161	-2.336	1.980
N	1.325	-1.988	-0.988
N	2.950	-0.872	0.400
H	1.029	-0.290	2.935
N	-0.428	-1.380	1.798
C	0.195	-0.238	2.252
C	0.765	-0.832	-1.385
H	-0.770	-1.756	-2.309
C	2.377	0.261	-0.019
N	-2.273	-1.723	0.232
C	-1.417	-0.975	0.935
N	-0.293	-0.870	-2.230
C	1.308	0.377	-0.910
H	3.391	1.841	0.986
N	2.676	1.555	0.335
N	-0.319	0.861	1.763
C	-1.326	0.419	0.926
H	-0.860	-0.038	-2.311
C	-3.057	-0.973	-0.541
H	-3.790	-1.506	-1.140
N	0.951	1.700	-1.093
C	1.785	2.363	-0.334
N	-3.048	0.361	-0.697
C	-2.183	1.087	0.031
H	1.807	3.437	-0.219
H	-1.307	2.898	0.152
N	-2.165	2.437	-0.112
H	-2.645	2.802	-0.921

adenine H-bond trimer	X	Y	Z
N	-1.386	2.809	0.006

C	-1.502	4.143	0.040
N	-0.538	5.061	0.071
C	0.671	4.488	0.050
C	0.959	3.124	0.006
C	-0.153	2.240	-0.006
N	2.330	2.919	-0.024
C	2.850	4.121	0.009
N	1.899	5.112	0.056
N	-0.050	0.910	-0.030
H	-2.520	4.523	0.047
H	3.906	4.345	0.001
H	2.053	6.108	0.084
H	-0.901	0.342	-0.067
H	0.851	0.435	0.048
N	4.037	0.462	-0.470
C	3.628	-0.764	-0.104
N	2.351	-0.949	0.296
C	1.957	-2.179	0.666
N	2.666	-3.306	0.694
C	3.923	-3.101	0.285
C	4.481	-1.890	-0.123
N	5.812	-2.043	-0.469
C	6.051	-3.314	-0.278
N	4.951	-4.011	0.178
H	0.923	-2.258	0.987
H	6.999	-3.803	-0.448
H	4.895	-4.994	0.398
H	3.432	1.275	-0.333
H	5.002	0.588	-0.725
N	-3.866	1.122	0.162
C	-3.795	-0.208	0.000
N	-2.591	-0.799	-0.169
C	-2.536	-2.130	-0.336
N	-3.537	-3.008	-0.355
C	-4.718	-2.403	-0.180
C	-4.938	-1.039	0.001
N	-6.286	-0.763	0.147
C	-6.866	-1.932	0.057
N	-5.974	-2.966	-0.140
H	-1.540	-2.542	-0.474
H	-7.929	-2.109	0.126
H	-6.184	-3.948	-0.240
H	-4.773	1.545	0.270
H	-3.024	1.707	0.110