

Supplementary Information

Near Field Response of Molecules Coupled with Plasmons at Atomistic Resolution

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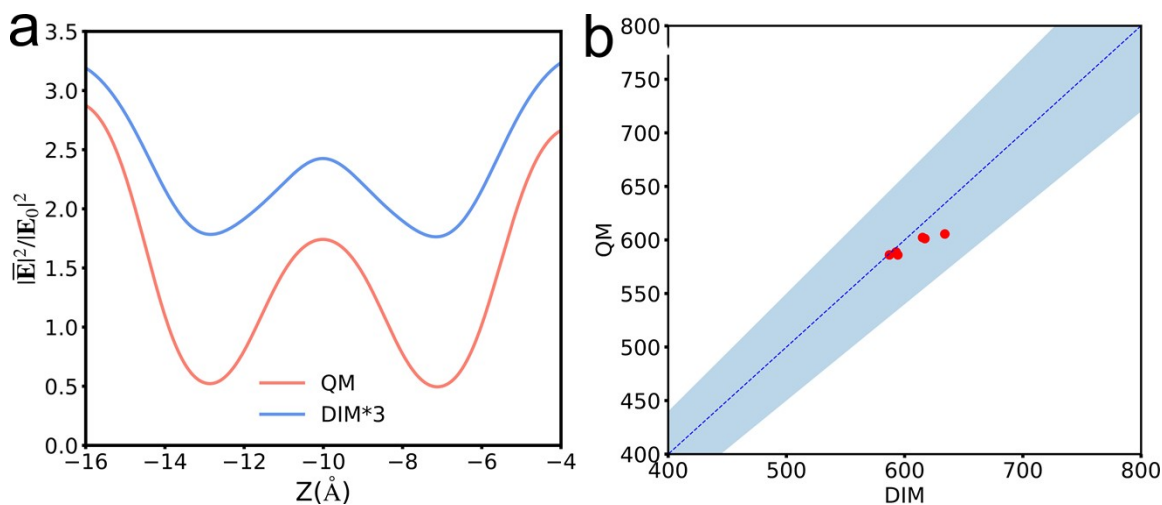


Fig. S1: Comparison of static polarizabilities of V^{+} viologen calculated by DIM and QM. The average static fields at bipyridine of an $n8 V^{+}$ viologen calculated by QM and DIM model, respectively (a). The zz-component polarizabilities of six V^{+} viologen molecules with 10% error in blue shading calculated by QM and DIM, respectively (b).

As shown in Fig. S1a, the average static near-fields of $n8 V^{+}$ viologen around the bipyridine moiety calculated by DIM, agree well with the results from QM calculations. According to Fig. S1b, the static zz-component polarizabilities of the V^{+} viologen obtained by DIM exhibit deviations from the QM results of less than 10%.

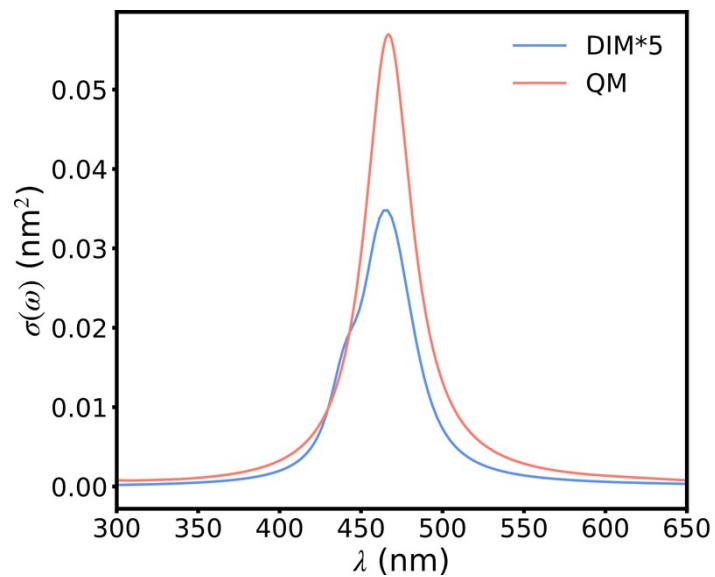


Fig. S2: The absorption peak of n8 V^{+•} viologen calculated by DIM and QM, respectively.

The maximum absorption of the n8 V^{+•} viologen, calculated both by DIM and QM is observed at approximately 465 nm (Fig. S2), indicating that the parameters employed in DIM model are capable of predicting the optical response characteristics of V^{+•} viologen molecules.

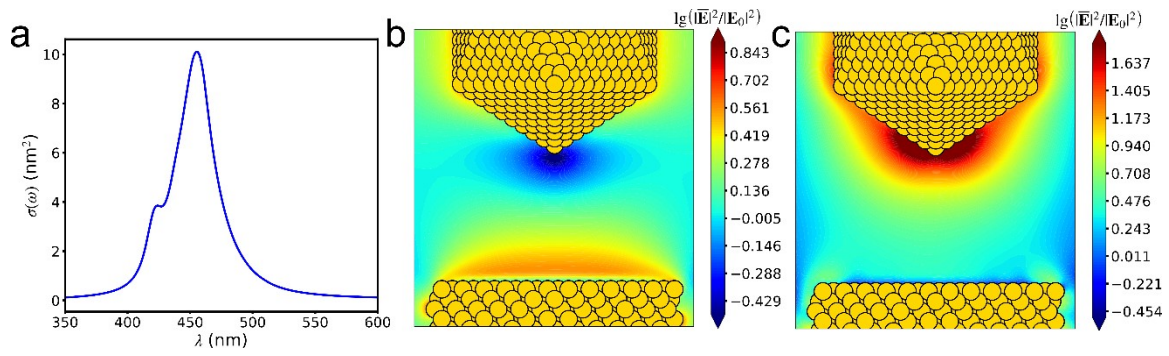


Fig. S3: Absorption spectra of the AuNC (a). The near-field distributions on the xz plane of the AuNC at the excitation wavelengths of 424 nm (b) and 456 nm (c), respectively.

The maximum absorption at 456 nm (Fig. S3a) is mainly attributed to the plasmon resonance of the Au tip (Fig. S3c). The weak absorption at 424 nm primarily arises from the response of the Au(111) surface (Fig. S3b).

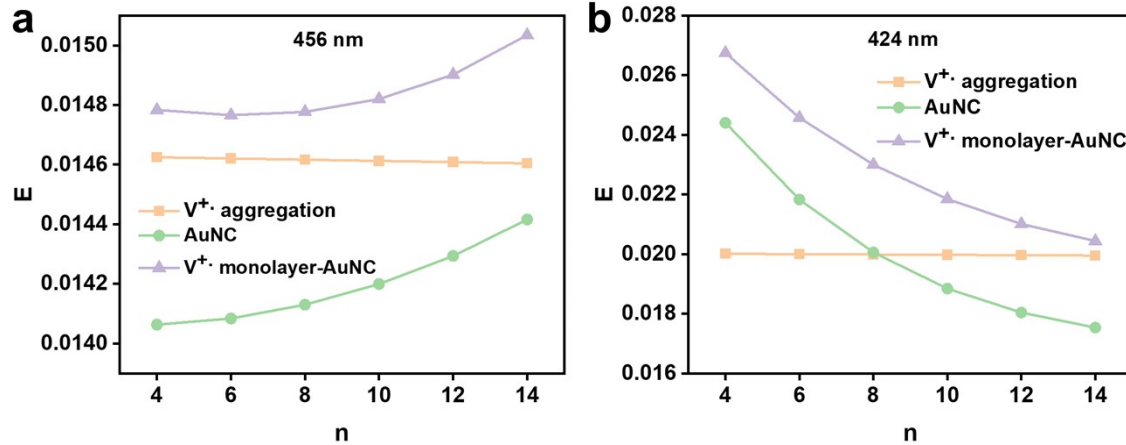


Fig. S4: The near-field within AuNC and free V^+ aggregation under 456 nm (a) and 424 nm (b) excitation simulated by FDTD.

For multi-molecular systems, FDTD simulations were performed in comparison with DIM results. Au icosahedron in DIM is simplified to an Au sphere and the Au (111) substrate to an Au slab. By adjusting the model size and permittivity constant, the longitudinal field strengths within the Au cavity were calculated at different excitation wavelengths. As shown in Figure. S4, at 456 nm excitation, a stronger near-field is observed near the Au sphere, while at 424 nm, the field is more intense near the Au slab, which are consistent with the DIM simulation results (Figure 5c blue and red curves). The molecular excitation properties were correlated with dielectric functions. To simplify the simulations, a dielectric function was employed, neglecting the exciton mode differences arising from distinct molecules. Upon embedding the molecular layers within the Au cavity, the significant near-field intensities within the cavity were observed, and the trend of the field strength distribution remained consistent with our DIM findings (Figure 5e blue and red curves).

Coordinates of all molecules:

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n4 monocationic viologen

S -2.62500000 -0.41600000 -24.07000000

C -2.72200000 -0.30000000 -22.23200000

C -1.30600000 -0.31200000 -21.65600000

C -1.29900000 -0.21700000 -20.12500000

C 0.13000000 -0.23300000 -19.56900000

N 0.17700000 -0.14100000 -18.09600000

C 0.13000000 -1.27100000 -17.32400000

C 0.11800000 -1.21100000 -15.96200000

C 0.14800000 0.03800000 -15.26500000

C 0.19400000 1.18900000 -16.11300000

C 0.20400000 1.07700000 -17.47200000

C 0.13900000 0.12900000 -13.83800000

C 0.08900000 -1.02200000 -12.99000000

C 0.07700000 -0.90800000 -11.63100000

N 0.10600000 0.30800000 -11.00700000

C 0.16000000 1.43600000 -11.77900000

C 0.17300000 1.37700000 -13.14100000

C 0.15200000 0.40000000 -9.53200000

C 1.57800000 0.37800000 -8.97300000

C 1.59200000 0.47600000 -7.44200000

C 3.00500000 0.45400000 -6.85100000

C 3.02100000 0.55100000 -5.32100000

C 4.43100000 0.52800000 -4.72200000

C 4.44400000 0.62700000 -3.19200000

C 5.85100000 0.60200000 -2.58700000

H -3.29600000 -1.14600000 -21.84900000
H -3.23400000 0.62500000 -21.95800000
H -0.73600000 0.52200000 -22.08000000
H -0.79800000 -1.23100000 -21.97000000
H -1.86200000 -1.05200000 -19.69400000
H -1.80100000 0.70200000 -19.80400000
H 0.64900000 -1.14900000 -19.85600000
H 0.71000000 0.60300000 -19.96700000
H -0.43400000 -0.43300000 -9.13700000
H -0.36600000 1.31900000 -9.24800000
H 2.14700000 1.21000000 -9.40400000
H 2.07700000 -0.54300000 -9.29200000
H 1.01000000 -0.35300000 -7.01900000
H 1.08300000 1.39600000 -7.13100000
H 3.58900000 1.28100000 -7.27400000
H 3.51400000 -0.46800000 -7.16100000
H 2.43500000 -0.27500000 -4.90000000
H 2.51300000 1.47400000 -5.01300000
H 5.02000000 1.35300000 -5.14300000
H 4.94000000 -0.39500000 -5.02900000
H 3.85500000 -0.19800000 -2.77200000
H 3.93500000 1.55000000 -2.88600000
C 5.86100000 0.70200000 -1.05800000
C 7.26700000 0.67500000 -0.44900000
C 7.27500000 0.77700000 1.08100000
C 8.68400000 0.74900000 1.68100000
H 6.44200000 1.42600000 -3.00700000
H 6.36000000 -0.32200000 -2.89200000
H 5.26900000 -0.12200000 -0.63800000

H 5.35200000 1.62600000 -0.75300000
H 7.86000000 1.49900000 -0.86800000
H 7.77600000 -0.24900000 -0.75200000
H 6.68400000 -0.04700000 1.50000000
H 6.76700000 1.69900000 1.38400000
H 9.28900000 1.58100000 1.30900000
H 9.20500000 -0.17900000 1.42500000
H 8.65400000 0.82200000 2.77200000
H 0.11000000 -2.21000000 -17.85900000
H 0.09000000 -2.15100000 -15.43100000
H 0.22700000 2.18800000 -15.70500000
H 0.24200000 1.94200000 -18.12000000
H 0.05400000 -2.02200000 -13.39800000
H 0.03700000 -1.77200000 -10.98200000
H 0.18300000 2.37600000 -11.24300000
H 0.20700000 2.31800000 -13.67200000

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n6 monocationic viologen

C -2.23300000 0.33500000 -22.54900000
C -2.33300000 0.37600000 -21.01800000
C -0.96200000 0.35400000 -20.33000000
C -1.06100000 0.37300000 -18.80000000
C 0.31900000 0.33800000 -18.13600000
N 0.24800000 0.34300000 -16.66000000
C 0.13000000 -0.82900000 -15.96300000
C -0.00500000 -0.85000000 -14.60700000
C -0.03700000 0.35600000 -13.83700000
C 0.09200000 1.55400000 -14.60800000

C 0.22300000 1.52100000 -15.96400000
C -0.18200000 0.36200000 -12.41400000
C -0.34300000 -0.83500000 -11.64800000
C -0.49000000 -0.80200000 -10.29300000
N -0.49400000 0.37500000 -9.59500000
C -0.33500000 1.54400000 -10.28700000
C -0.18600000 1.56500000 -11.64200000
C -0.60000000 0.37600000 -8.12000000
C 0.75700000 0.25800000 -7.42000000
C 0.61200000 0.26100000 -5.89300000
C 1.95300000 0.13700000 -5.16000000
C 1.80800000 0.14100000 -3.63400000
C 3.14300000 0.01400000 -2.89300000
C 2.99500000 0.02100000 -1.36800000
C 4.32800000 -0.10700000 -0.62200000
H -2.92600000 -0.47900000 -20.67000000
H -2.88000000 1.27600000 -20.70800000
H -0.37200000 1.21300000 -20.67000000
H -0.41300000 -0.54100000 -20.64900000
H -1.64800000 -0.48500000 -18.45500000
H -1.59600000 1.27100000 -18.47200000
H 0.87100000 -0.55700000 -18.43200000
H 0.91900000 1.20000000 -18.43600000
H -1.25600000 -0.45200000 -7.84100000
H -1.10800000 1.29900000 -7.83300000
H 1.39900000 1.08800000 -7.73500000
H 1.25300000 -0.66300000 -7.74500000
H -0.04500000 -0.56400000 -5.58800000
H 0.11100000 1.18400000 -5.57700000

H 2.6100000 0.9600000 -5.4650000
H 2.4530000 -0.7880000 -5.4750000
H 1.1460000 -0.6800000 -3.3310000
H 1.3070000 1.0660000 -3.3210000
H 3.8060000 0.8350000 -3.1960000
H 3.6440000 -0.9110000 -3.2040000
H 2.3310000 -0.7990000 -1.0650000
H 2.4950000 0.9470000 -1.0570000
C 4.1790000 -0.0980000 0.9040000
C 5.5150000 -0.2230000 1.6420000
H -1.6440000 1.1900000 -22.9000000
H 4.9930000 0.7110000 -0.9240000
H 4.8280000 -1.0350000 -0.9290000
H 3.5170000 -0.9180000 1.2060000
H 3.6770000 0.8270000 1.2110000
H 6.1860000 0.6040000 1.3890000
H 6.0260000 -1.1540000 1.3810000
H 0.1580000 -1.7350000 -16.5520000
H -0.0800000 -1.8190000 -14.1370000
H 0.0880000 2.5270000 -14.1390000
H 0.3190000 2.4220000 -16.5550000
H -0.3640000 -1.8070000 -12.1190000
H -0.6170000 -1.7010000 -9.7060000
H -0.3400000 2.4490000 -9.6940000
H -0.0720000 2.5330000 -12.1070000
H 5.3740000 -0.2150000 2.7260000
C -3.6020000 0.3510000 -23.2280000
S -3.5430000 0.1900000 -25.0600000
H -1.6900000 -0.5630000 -22.8620000

H -4.20000000 -0.50600000 -22.90600000

H -4.16400000 1.25100000 -22.96900000

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n8 monocationic viologen

C -3.49900000 0.10000000 -21.60600000

C -3.57100000 0.23000000 -20.07800000

C -2.21000000 0.04700000 -19.39700000

C -2.27300000 0.15800000 -17.87000000

C -0.90900000 -0.03000000 -17.19600000

C -0.97500000 0.07200000 -15.66700000

C 0.40100000 -0.12100000 -15.02200000

N 0.36600000 -0.03400000 -13.54700000

C 0.13000000 -1.14600000 -12.78600000

C 0.03700000 -1.08300000 -11.42800000

C 0.17600000 0.15400000 -10.72300000

C 0.41700000 1.28900000 -11.55900000

C 0.50200000 1.17200000 -12.91500000

C 0.08400000 0.24800000 -9.29800000

C -0.14700000 -0.88900000 -8.46100000

C -0.23300000 -0.77200000 -7.10600000

N -0.10900000 0.43500000 -6.47400000

C 0.11900000 1.54900000 -7.23600000

C 0.21300000 1.48600000 -8.59400000

C -0.14600000 0.52400000 -4.99900000

C 1.23200000 0.35500000 -4.35300000

C 1.16200000 0.45100000 -2.82400000

C 2.52800000 0.28800000 -2.14800000

C 2.46000000 0.38300000 -0.61900000

C 3.82300000 0.22300000 0.06300000
C 3.75600000 0.31800000 1.59100000
C 5.12000000 0.15700000 2.26700000
C -4.86000000 0.28300000 -22.27700000
H -3.09600000 -0.88000000 -21.87700000
H -2.79700000 0.84300000 -22.00300000
H -4.27600000 -0.51300000 -19.68400000
H -3.98200000 1.21200000 -19.81200000
H -1.50700000 0.79500000 -19.78600000
H -1.79700000 -0.93100000 -19.67300000
H -2.97600000 -0.59000000 -17.48100000
H -2.68300000 1.13700000 -17.59200000
H -0.20800000 0.72000000 -17.58100000
H -0.49800000 -1.00700000 -17.47800000
H -1.66600000 -0.68200000 -15.27300000
H -1.37700000 1.04800000 -15.37500000
H 1.10600000 0.63400000 -15.37600000
H 0.81900000 -1.09700000 -15.28000000
H 0.02800000 -2.07700000 -13.32700000
H -0.14300000 -2.01100000 -10.90500000
H 0.54700000 2.27700000 -11.14400000
H 0.68800000 2.02400000 -13.55500000
H -0.26500000 -1.87900000 -8.87600000
H -0.41000000 -1.62500000 -6.46500000
H 0.21100000 2.48100000 -6.69500000
H 0.38300000 2.41600000 -9.11700000
H -0.83900000 -0.24300000 -4.64500000
H -0.58100000 1.49200000 -4.74300000
H 1.91000000 1.12200000 -4.74400000

H 1.65200000 -0.61400000 -4.64600000
H 0.47500000 -0.31400000 -2.44200000
H 0.73000000 1.41900000 -2.53900000
H 3.21500000 1.05400000 -2.52900000
H 2.96000000 -0.67900000 -2.43300000
H 1.77300000 -0.38300000 -0.23900000
H 2.02500000 1.35000000 -0.33600000
H 4.51100000 0.98900000 -0.31600000
H 4.25900000 -0.74400000 -0.22000000
H 3.06800000 -0.44800000 1.97000000
H 3.32000000 1.28300000 1.87400000
H 5.82000000 0.93100000 1.93500000
H 5.56700000 -0.81400000 2.03200000
H 5.03700000 0.23000000 3.35400000
S -4.84900000 0.03400000 -24.10200000
H -5.28300000 1.26500000 -22.05800000
H -5.57100000 -0.46400000 -21.91400000

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n10 monocationic viologen

C -1.73200000 0.01700000 -17.80900000
C -1.89000000 0.13000000 -16.28800000
C -0.57900000 -0.10300000 -15.52700000
C -0.73900000 0.00800000 -14.00600000
C 0.58500000 -0.23200000 -13.27400000
N 0.46100000 -0.13200000 -11.80500000
C 0.13000000 -1.22800000 -11.05500000
C -0.04400000 -1.15100000 -9.70500000
C 0.10500000 0.08400000 -8.99900000

C 0.44600000 1.20100000 -9.82400000
C 0.60900000 1.07100000 -11.17100000
C -0.06900000 0.19200000 -7.58300000
C -0.40700000 -0.92600000 -6.75700000
C -0.56800000 -0.79700000 -5.41000000
N -0.42200000 0.40700000 -4.77600000
C -0.09500000 1.50300000 -5.52700000
C 0.07700000 1.42700000 -6.87700000
C -0.54100000 0.50600000 -3.30600000
C 0.78800000 0.27200000 -2.58100000
C 0.63600000 0.38100000 -1.05900000
C 1.95200000 0.15100000 -0.30600000
H -2.64100000 -0.59200000 -15.94600000
H -2.28400000 1.12300000 -16.03700000
H 0.17100000 0.62100000 -15.86800000
H -0.18600000 -1.09500000 -15.78100000
H -1.48200000 -0.71800000 -13.65600000
H -1.12200000 1.00000000 -13.74300000
H 0.98000000 -1.22500000 -13.50000000
H 1.33900000 0.49300000 -13.58700000
H -1.29000000 -0.22200000 -2.99100000
H -0.93900000 1.49700000 -3.07900000
H 1.52600000 1.00100000 -2.93400000
H 1.17400000 -0.71800000 -2.84700000
H -0.11000000 -0.34600000 -0.71400000
H 0.24000000 1.37100000 -0.80200000
H 2.69900000 0.87700000 -0.65000000
H -0.98000000 0.74000000 -18.15000000
H 0.02200000 -2.15700000 -11.59700000

H -0.29500000 -2.06700000 -9.19100000
H 0.59100000 2.18600000 -9.40600000
H 0.87000000 1.91000000 -11.80300000
H -0.54900000 -1.91200000 -7.17500000
H -0.82500000 -1.63600000 -4.77800000
H 0.01100000 2.43300000 -4.98500000
H 0.32400000 2.34400000 -7.39200000
C -3.03900000 0.24500000 -18.57700000
H -1.33500000 -0.97500000 -18.05800000
H -3.43500000 1.23800000 -18.33200000
H -3.79100000 -0.47600000 -18.23400000
C -2.87800000 0.12300000 -20.09600000
C -4.18400000 0.34600000 -20.86900000
C -4.01500000 0.20800000 -22.38800000
C -5.32100000 0.43400000 -23.14800000
S -5.19700000 0.18200000 -24.96900000
H 2.34900000 -0.84000000 -0.56300000
H -2.48000000 -0.87000000 -20.34000000
H -2.12600000 0.84500000 -20.44000000
H -4.57900000 1.34200000 -20.63300000
H -4.93700000 -0.37200000 -20.52100000
H -3.62800000 -0.78700000 -22.63000000
H -3.26300000 0.92400000 -22.73800000
H -5.72400000 1.43000000 -22.95800000
H -6.07900000 -0.28800000 -22.83300000
C 1.80600000 0.25800000 1.21600000
C 3.12200000 0.02900000 1.96300000
H 1.05900000 -0.46800000 1.55900000
H 1.40800000 1.24800000 1.47100000

H 3.87800000 0.76300000 1.66800000
H 3.52700000 -0.96600000 1.75700000
H 2.98400000 0.11300000 3.04300000

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n12 monocationic viologen

C -2.79500000 0.29400000 -15.32100000
C -2.76200000 0.31200000 -13.78800000
C -1.34000000 0.28300000 -13.21500000
C -1.31200000 0.30200000 -11.68100000
C 0.11900000 0.27100000 -11.13600000
N 0.17600000 0.29100000 -9.65900000
C 0.13000000 -0.87300000 -8.94100000
C 0.12300000 -0.87900000 -7.57800000
C 0.15600000 0.33500000 -6.82200000
C 0.19900000 1.52500000 -7.61400000
C 0.20500000 1.47700000 -8.97600000
C 0.15200000 0.35600000 -5.39200000
C 0.12100000 -0.83300000 -4.60000000
C 0.11700000 -0.78700000 -3.23700000
N 0.13500000 0.40000000 -2.55500000
C 0.16900000 1.56500000 -3.27300000
C 0.17400000 1.57000000 -4.63600000
C 0.19400000 0.42100000 -1.07900000
C 1.62600000 0.40300000 -0.53600000
H -3.32500000 -0.54800000 -13.40300000
H -3.28200000 1.20600000 -13.42400000
H -0.77700000 1.14200000 -13.60000000
H -0.82000000 -0.61200000 -13.57800000

H -1.86600000 -0.55700000 -11.28800000
H -1.81900000 1.20000000 -11.31000000
H 0.64400000 -0.62700000 -11.46900000
H 0.69300000 1.12900000 -11.49500000
H -0.37100000 -0.44100000 -0.71800000
H -0.33800000 1.31500000 -0.74500000
H -2.23200000 1.15400000 -15.70500000
H 0.10600000 -1.78500000 -9.52100000
H 0.09400000 -1.84300000 -7.09300000
H 0.23400000 2.50300000 -7.15800000
H 0.24000000 2.37200000 -9.58400000
H 0.09800000 -1.81300000 -5.05600000
H 0.09200000 -1.68100000 -2.63000000
H 0.18300000 2.47700000 -2.69300000
H 0.19100000 2.53600000 -5.12200000
C -4.21300000 0.31800000 -15.90000000
H -2.27100000 -0.60000000 -15.68400000
H -4.73800000 1.21200000 -15.53800000
H -4.77600000 -0.54200000 -15.51500000
C -4.24600000 0.29900000 -17.43200000
C -5.66300000 0.31900000 -18.01500000
C -5.69400000 0.29500000 -19.54700000
C -7.11000000 0.31100000 -20.13000000
H -3.72000000 -0.59300000 -17.79400000
H -3.68500000 1.15900000 -17.81700000
H -6.19000000 1.21300000 -17.65600000
H -6.22600000 -0.54000000 -17.62800000
H -5.16500000 -0.59800000 -19.90400000
H -5.13300000 1.15500000 -19.93300000

H -7.63800000 1.20700000 -19.77900000
H -7.67300000 -0.54700000 -19.74000000
C -7.13100000 0.27400000 -21.66500000
C -8.54900000 0.29300000 -22.23500000
S -8.63700000 0.15300000 -24.07000000
H 2.17300000 1.26500000 -0.93200000
H -6.61400000 -0.62200000 -22.02200000
H -6.57100000 1.13100000 -22.05800000
H -9.08900000 1.18900000 -21.92500000
H -9.11900000 -0.56800000 -21.87800000
C 1.66000000 0.42400000 0.99700000
C 3.08500000 0.40800000 1.55700000
H 2.14100000 -0.49100000 -0.90600000
H 1.10400000 -0.43600000 1.38500000
H 1.13500000 1.31600000 1.35900000
H 3.65600000 1.27700000 1.21600000
H 3.62400000 -0.49000000 1.24200000
H 3.07600000 0.42400000 2.64800000

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n14 monocationic viologen

C -1.72100000 -0.18500000 -13.05400000
C -1.88200000 -0.08900000 -11.53200000
C -0.56600000 -0.28200000 -10.77000000
C -0.73600000 -0.18200000 -9.24900000
C 0.59100000 -0.38000000 -8.50900000
N 0.45000000 -0.28300000 -7.04000000
C 0.13000000 -1.38400000 -6.29400000
C -0.06700000 -1.31000000 -4.94700000

C 0.04400000 -0.07200000 -4.24000000
C 0.37800000 1.05100000 -5.06100000
C 0.56500000 0.92300000 -6.40500000
C -0.15900000 0.03400000 -2.82900000
C -0.48800000 -1.08900000 -2.00700000
C -0.67600000 -0.96200000 -0.66300000
N -0.56800000 0.24700000 -0.02900000
C -0.25200000 1.35000000 -0.77600000
C -0.05400000 1.27400000 -2.12200000
C -0.71300000 0.34600000 1.43900000
C 0.60900000 0.14400000 2.17800000
H -2.60800000 -0.84000000 -11.19600000
H -2.31000000 0.88700000 -11.27400000
H 0.16100000 0.46800000 -11.10600000
H -0.13800000 -1.26000000 -11.02600000
H -1.45600000 -0.93300000 -8.90500000
H -1.15300000 0.79600000 -8.98500000
H 1.01900000 -1.36000000 -8.73000000
H 1.32300000 0.36900000 -8.81500000
H -1.45000000 -0.40000000 1.74200000
H -1.13900000 1.32600000 1.65600000
H -0.99600000 0.56700000 -13.38900000
H 0.04900000 -2.31600000 -6.83700000
H -0.30700000 -2.23100000 -4.43600000
H 0.49800000 2.03900000 -4.64100000
H 0.81900000 1.76600000 -7.03300000
H -0.60200000 -2.07900000 -2.42500000
H -0.92700000 -1.80600000 -0.03400000
H -0.17700000 2.28300000 -0.23500000

H 0.18100000 2.19600000 -2.63500000
C -3.03700000 0.00500000 -13.81600000
H -1.29000000 -1.16100000 -13.31200000
H -3.46800000 0.98000000 -13.55700000
H -3.76200000 -0.74700000 -13.48000000
C -2.88000000 -0.09000000 -15.33800000
C -4.19600000 0.09700000 -16.10000000
C -4.04100000 0.00100000 -17.62200000
C -5.35800000 0.18800000 -18.38400000
H -2.44700000 -1.06500000 -15.59700000
H -2.15600000 0.66300000 -15.67500000
H -4.62800000 1.07300000 -15.84200000
H -4.92000000 -0.65500000 -15.76200000
H -3.60900000 -0.97500000 -17.88100000
H -3.31700000 0.75300000 -17.96100000
H -5.78900000 1.16400000 -18.12700000
H -6.08300000 -0.56300000 -18.04200000
C -5.20500000 0.08600000 -19.90500000
C -6.52300000 0.27200000 -20.66500000
H 1.34400000 0.89900000 1.89000000
H -4.77500000 -0.89100000 -20.16100000
H -4.48000000 0.83600000 -20.24700000
H -6.95000000 1.25100000 -20.41400000
H -7.24800000 -0.47500000 -20.32100000
H 1.03100000 -0.84300000 1.97600000
C -6.36300000 0.15900000 -22.18800000
C -7.68300000 0.35200000 -22.93200000
S -7.57100000 0.13400000 -24.75900000
H 0.44200000 0.22800000 3.25400000

H -5.94600000 -0.82000000 -22.44700000

H -5.63900000 0.90500000 -22.53500000

H -8.11900000 1.33100000 -22.72200000

H -8.41300000 -0.40000000 -22.62200000