Supporting Information: Quantum Embedding Method for the Simulation of Strongly Correlated Systems on Quantum Computers

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1 C-N bond Dissociation in $CH_3CH_2CH_2CN$ Calculated with Different WF-in-HF Methods

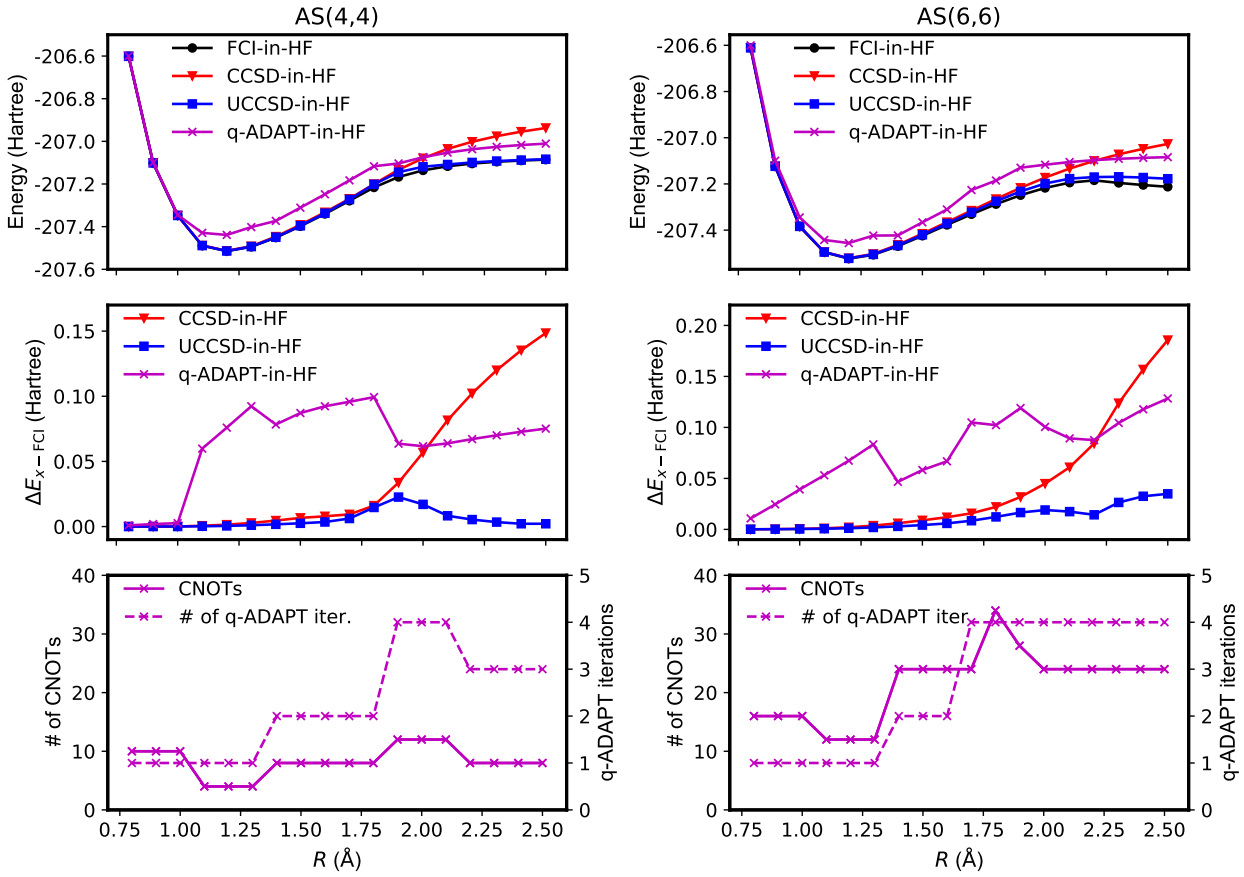

Figure S1: Potential energy surface (upper panels) for the triple C-N bond dissociation in $CH_3CH_2CH_2CN$ calculated with different WF-in-HF/STO-3G methods and two active spaces, AS(4,4) (left column) and AS(6,6) (right column). The middle panels show the error for a respective embedding method relative to the reference FCI-in-HF method. The lowest panels indicate the number of CNOT gates (solid magenta line) and number of q-ADAPT-in-HF iterations (dashed magenta line).

The upper two panels of Fig. S1 show the potential energy surface for the triple C-N bond dissociation in butyronitrile calculated with different WF-in-HF methods and two active spaces, AS(4,4) (left column) and AS(6,6) (right column). The reference values were calculated with the FCI-in-HF method (black line) which shows a proper behavior for the triple bond dissociation. In case of the CCSD-in-HF method (red line), the energy curve remains in excellent agreement with the reference curve for values of $R < 1.7$ Å, however,

for larger values of R the energy starts to deviate rapidly. The UCCSD-in-HF method (blue line) displays much lower discrepancy in energy with respect to the FCI-in-HF curve across the whole dissociation curve. The middle two panels show the error of a given method with respect to the reference FCI-in-HF method. The q-ADAPT-in-HF (magenta line) remains mostly parallel to the reference FCI-in-HF curve, thus, properly describing a correct dissociation of the triple C-N bond for both active spaces. The lowest panels show the number of CNOT gates (solid magenta line) and number of iterations (dashed magenta line) for the q-ADAPT-in-HF method for a maximum number of four iterations while targeting a maximum operator gradient of 1e$-$4 and a maximum energy expectation value threshold of 100 mHartree. For AS(4,4) the average number of CNOT gates is 8, whereas for AS(6,6), the average number of the CNOT gates is 21.

2 C-N bond Dissociation in $CH_3CH_2CH_2CN$ Calculated with the PBE-in-PBE and FCI-in-PBE Methods

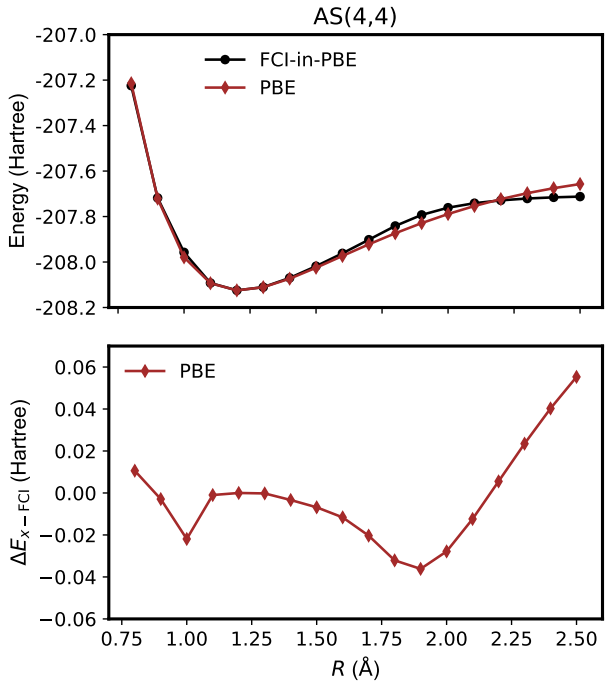

Figure S2: Potential energy surface (upper panel) for the triple C-N bond dissociation in $CH_3CH_2CH_2CN$ calculated with FCI-in-PBE (black line) method for AS(4,4), and with the PBE (brown line) method. Note that the PBE method is equivalent to the PBE-in-PBE method. The PBE curve was shifted such that its minimum matches the minimum of the FCI-in-PBE method. The lower panel shows the error for the PBE method relative to the reference FCI-in-PBE method.

The upper panel of Fig. S2 shows the potential energy surface for the triple C-N bond dissociation in butyronitrile calculated with FCI-in-PBE method (black line) for AS(4,4), and with the PBE (brown line) method. Note that due to the exactness of the projection-based embedding approach, the PBE-in-PBE and PBE methods are equivalent. The PBE curve was shifted such that its minimum equals the FCI-in-PBE minimum. The lower panel shows the error of the PBE method with respect to the FCI-in-PBE method. As evident from both plots, the PBE method fails to properly describe the triple bond breaking due to

its use of a single Slater determinant.

3 C-N bond Dissociation in $CH_3CH_2CH_2CN$ Calculated with q-ADAPT-in-PBE Method with Different Thresholds

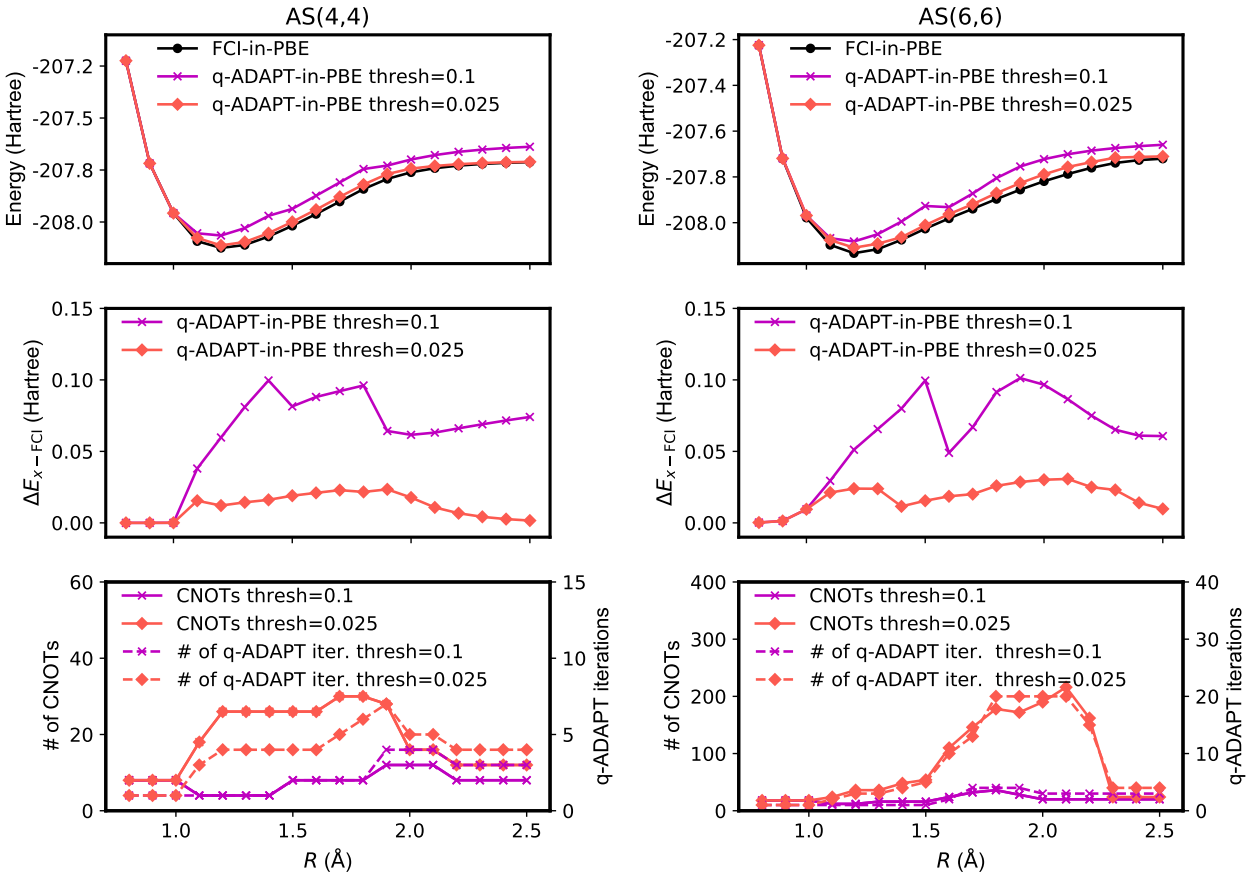

Figure S3: Potential energy surface (upper panels) for the triple C-N bond dissociation in $CH_3CH_2CH_2CN$ calculated with FCI-in-PBE/STO-3G method (black line), and with the q-ADAPT-in-PBE method using two different thresholds for two active spaces, AS(4,4) (left column) and AS(6,6) (right column). The middle panels show the error for the q-ADAPT-in-PBE method with different thresholds relative to the reference FCI-in-PBE method. The lowest panels indicate the number of CNOT gates and number of q-ADAPT-in-PBE iterations with two different thresholds.

The upper panels of Fig. S3 show the potential energy surface for the triple C-N bond dissociation in butyronitrile calculated with FCI-in-PBE method (black line), and with the q-ADAPT-in-PBE method using two different convergence threshold criteria. The first one

denoted as thresh=0.1 (magenta line) corresponds to the q-ADAPT-in-PBE results obtained for a maximum number of four iterations while targeting a maximum operator gradient of 1e−4 and a maximum energy expectation value threshold of 100 mHartree for both active spaces (corresponding to the thresholds used for the results of the main manuscript). In case of the second one denoted as thresh=0.025 (orange line), for the AS(4,4) case (left column), the q-ADAPT-in-PBE results are obtained for a maximum of seven iterations at an operator gradient threshold of 1e−4 and an energy expectation value threshold of 25 mHartree. The AS(6,6) case (right column), the q-ADAPT-in-VQE results are obtained for a maximum of twenty iterations at an operator gradient threshold of 1e−4 and an energy energy expectation value threshold of 25 mHartree. As can be seen from the upper and middle panels, the orange curve shows little deviation relative to the FCI-in-PBE curve. The lowest panels show the number of CNOT gates (solid lines) and number of iterations (dashed lines) for q-ADAPT-in-PBE method with two different threshold criteria. For AS(4,4) the average number of CNOT gates is 8 and 19 for two different thresholds, whereas for AS(6,6), the average number of the CNOT gates is 21 and 83, respectively.

4 C-N bond Dissociation in $CH_3CH_2CH_2CN$ Calculated with f-ADAPT-in-PBE Method

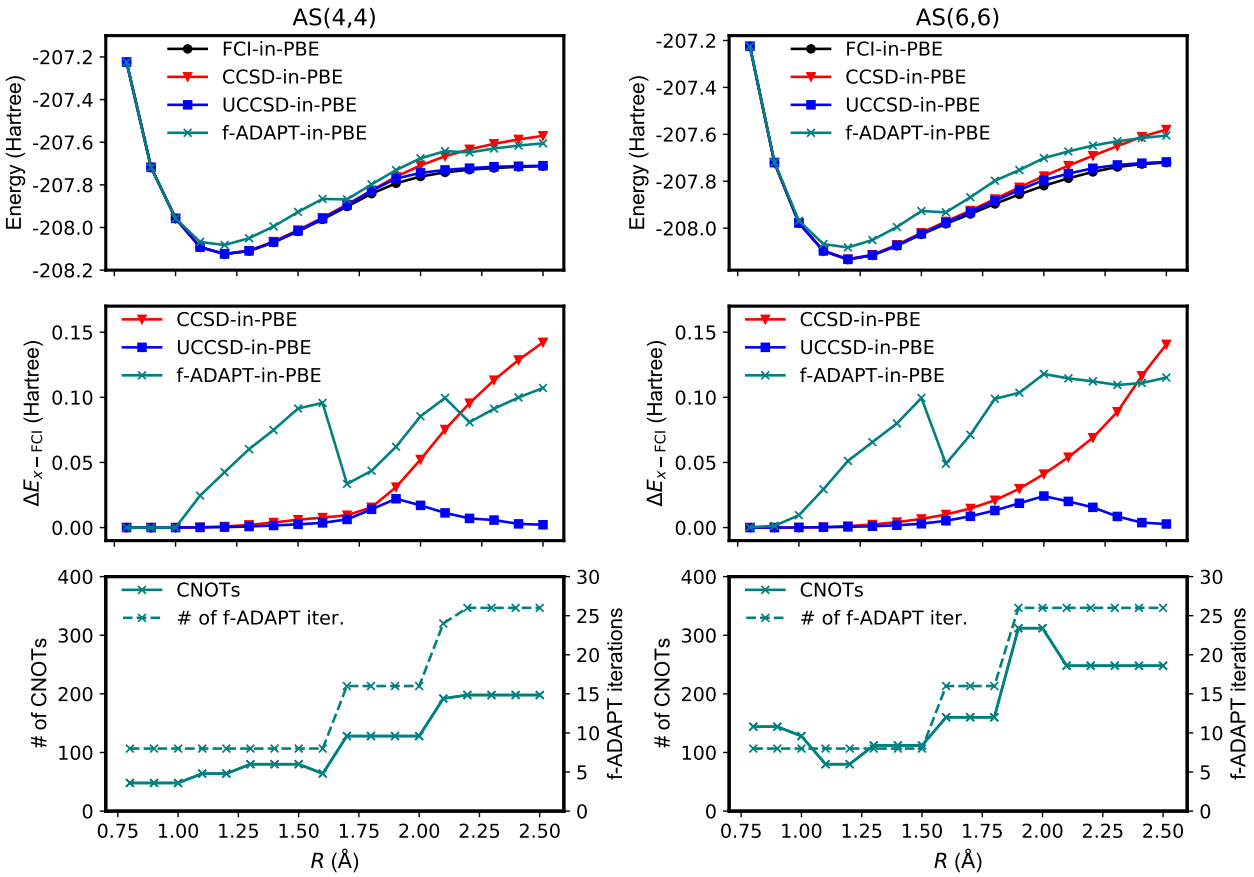

Figure S4: Potential energy surface (upper panels) for the triple C-N bond dissociation in $CH_3CH_2CH_2CN$ calculated with different WF-in-PBE/STO-3G methods and two active spaces, AS(4,4) (left column) and AS(6,6) (right column). The middle panels show the error for a respective embedding method relative to the reference FCI-in-PBE method. The lowest panels indicate the number of CNOT gates and number of f-ADAPT-in-PBE iterations.

The upper two panels of Fig. S4 show the potential energy surface for the triple C-N bond dissociation in butyronitrile calculated with the f-ADAPT-in-PBE method (teal line) and two active spaces, AS(4,4) (left column) and AS(6,6) (right column). The results obtained with the FCI-in-PBE (black line) and CCSD-in-PBE (blue line) methods are included for comparison. The middle two panels show the error of a given method with respect to the reference FCI-in-PBE method. For selected set of threshold parameters (maximum

of four iterations while targeting a maximum operator gradient of 1e−4 and a maximum energy expectation value threshold of 100 mHartree), the f-ADAPT-in-HF (teal line) shows qualitatively better behavior for dissociation of the triple triple C-N bond compared with the CCSD-in-PBE method. The lowest panels show the number of CNOT gates (solid teal line) and number of iterations (dashed teal line) for f-ADAPT-in-PBE method. For AS(4,4) the average number of CNOT gates is 115, whereas for AS(6,6), the average number of the CNOT gates is 181. For the selected set of threshold parameters, the q-ADAPT-in-PBE method used in the main manuscript exhibits much better agreement relative to the FCI-in-PBE method then the f-ADAPT-in-PBE method.

5 Shifted C-N bond Dissociation in $CH_3CH_2CH_2CN$ Curve Calculated with ibm_cairo-in-PBE Method

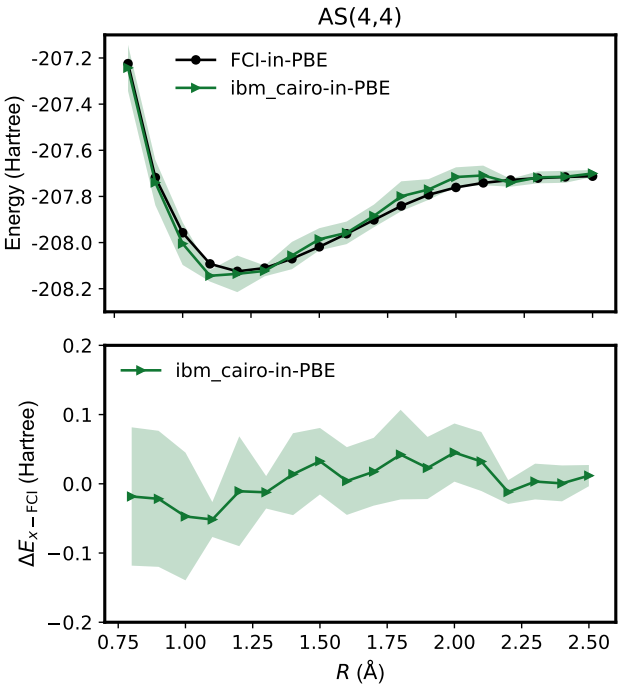

Figure S5: Potential energy surface (upper panel) for the triple C-N bond dissociation in $CH_3CH_2CH_2CN$ calculated with the FCI-in-PBE (black line) method and ibm_cairo-in-PBE (dark green line) method shifted down by 0.12 Hartree relative to the original values. The shaded green area indicates the standard deviation between 10 independent experiment repetitions and the dark green line corresponds to the average value. The lower panel shows the error for the shifted ibm_cairo-in-PBE method relative to the reference FCI-in-PBE method.

The upper panel of Fig. S5 shows the potential energy surface for the triple C-N bond dissociation in butyronitrile calculated with FCI-in-PBE (black line) and with ibm_cairo-in-PBE (dark green line) methods for AS(4,4). The ibm_cairo-in-PBE values are shifted down by 120 mHartree from its original values. The lower panel shows the error for the shifted ibm_cairo-in-PBE method relative to the reference FCI-in-PBE method. The shaded green area in both panels indicates the standard deviation between 10 independent experiment repetitions.