

SUPPORTING INFORMATION

Table S1 The numbers of VSCF configurations selected for the P and Q spaces (N_P and N_Q) and the QDPT2 frequencies calculated by different values of A_{th} , B_{th} , and N_{gen} for the three Fermi polyads in CO₂.

	$N_{\text{gen}}^{\text{a}}$			A_{th}^{b}				B_{th}^{c}			
	1	2	3	0.3	0.2	0.1	0.05	0.15	0.1	0.05	0.01
N_P	3	3	3	3	3	3	3	3	3	3	3
N_Q	140	140	140	140	140	140	140	140	140	140	140
ν_1	1289.2	1289.2	1289.2	1289.2	1289.2	1289.2	1289.2	1289.2	1289.2	1289.2	1289.2
$2\nu_2^0$	1391.6	1391.6	1391.6	1391.6	1391.6	1391.6	1391.6	1391.6	1391.6	1391.6	1391.6
$2\nu_2^2$	1342.1	1342.1	1342.1	1342.1	1342.1	1342.1	1342.1	1342.1	1342.1	1342.1	1342.1
N_P	3	3	3	3	3	3	3	3	3	3	3
N_Q	162	162	162	162	162	162	162	162	162	162	162
$\nu_1\nu_2^1$	1938.6	1938.6	1938.6	1938.6	1938.6	1938.6	1938.6	1938.6	1938.6	1938.6	1938.6
$3\nu_2^1$	2015.4	2015.4	2015.4	2015.4	2015.4	2015.4	2015.4	2015.4	2015.4	2015.4	2015.4
$3\nu_2^3$	2085.3	2085.3	2085.3	2085.3	2085.3	2085.3	2085.3	2085.3	2085.3	2085.3	2085.3
N_P	5	6	6	6	6	6	6	5	6	6	6
N_Q	248	281	281	281	281	281	281	255	281	281	281
$\nu_1 2\nu_2^0$	2479.5	2555.6	2555.6	2555.6	2555.6	2555.6	2555.6	2420.8	2555.6	2555.6	2555.6
$2\nu_1$	2577.9	2594.1	2594.1	2594.1	2594.1	2594.1	2594.1	2594.1	2594.1	2594.1	2594.1
$4\nu_2^0$	2680.0	2677.6	2677.6	2677.6	2677.6	2677.6	2677.6	2675.8	2677.6	2677.6	2677.6
$\nu_1 2\nu_2^2$	2693.0	2690.1	2690.1	2690.1	2690.1	2690.1	2690.1	2735.3	2690.1	2690.1	2690.1
$4\nu_2^2$	2795.2	2776.3	2776.3	2776.3	2776.3	2776.3	2776.3	2776.3	2776.3	2776.3	2776.3
$4\nu_2^4$	-	2809.3	2809.3	2809.3	2809.3	2809.3	2809.3	-	2809.3	2809.3	2809.3

^a. $A_{\text{th}} = 0.1$ and $B_{\text{th}} = 0.05$. ^b. $N_{\text{gen}} = 3$ and $B_{\text{th}} = 0.05$. ^c. $N_{\text{gen}} = 3$ and $A_{\text{th}} = 0.1$.

Table S2 The numbers of VSCF configurations selected for the P and Q spaces (N_P and N_Q) and the QDPT2 frequencies calculated by different values of A_{th} , B_{th} , and N_{gen} for the three Fermi polyads in H_2CO .

	$N_{\text{gen}}^{\text{a}}$			A_{th}^{b}				B_{th}^{c}			
	1	2	3	0.3	0.2	0.1	0.05	0.15	0.1	0.05	0.01
N_P	3	3	3	3	3	3	3	2	3	3	3
N_Q	555	555	555	555	555	555	555	447	555	555	555
ν_5	2704.0	2704.0	2704.0	2704.0	2704.0	2704.0	2704.0	-	2704.0	2704.0	2704.0
$\nu_3\nu_6$	2841.1	2841.1	2841.1	2841.1	2841.1	2841.1	2841.1	2813.4	2841.1	2841.1	2841.1
N_P	3	4	4	3	3	4	4	3	3	4	7
N_Q	631	789	789	631	631	789	789	631	631	789	1093
$\nu_5\nu_6$	3919.7	3921.9	3921.9	3919.7	3919.7	3921.9	3921.9	3919.7	3919.7	3921.9	3922.3
$\nu_32\nu_6$	4088.6	4081.7	4081.7	4088.6	4088.6	4081.7	4081.7	4088.6	4088.6	4081.7	4082.4
N_P	4	7	9	6	7	9	10	7	7	9	10
N_Q	814	1245	1435	1129	1245	1435	1565	1292	1292	1435	1491
$\nu_3\nu_5\nu_6$	5382.0	5329.2	5329.5	5335.9	5329.2	5329.5	5329.6	5335.9	5335.9	5329.5	5329.6
$2\nu_32\nu_6$	-	5473.1	5473.1	-	5473.1	5473.1	5473.1	-	-	5473.1	5473.5
$2\nu_1$	5515.0	5538.5	5535.6	5516.8	5538.5	5535.6	5535.5	5516.7	5516.7	5535.6	5535.2
$2\nu_5$	5641.7	5658.2	5653.8	5657.6	5658.2	5653.8	5653.8	5658.3	5658.3	5653.8	5653.8

^a. $A_{\text{th}} = 0.1$ and $B_{\text{th}} = 0.05$.

^b. $N_{\text{gen}} = 3$ and $B_{\text{th}} = 0.05$.

^c. $N_{\text{gen}} = 3$ and $A_{\text{th}} = 0.1$.