

**SUPPLEMENTARY INFORMATION**

**to**

**First-Principles Calculation of Electron**

**Spin-Rotation Tensors**

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Table 1: Computed (all-electron CCSD/aug-cc-pCVTZ) and experimental structure of ( $XY_{\{2,3\}}$  or  $XYZ$ ) radicals

| Radical and state            | Structure    | $r_{XY}/\text{\AA}$ | $r_{YZ}/\text{\AA}$ | $\theta_{XYZ}/\text{degree}$ |
|------------------------------|--------------|---------------------|---------------------|------------------------------|
| $\text{NH}_2 \tilde{X}^2B_1$ | Comp. $r_e$  | 1.0236              |                     | 103.20                       |
|                              | Exp. $r_0^1$ | 1.0245(37)          |                     | 103.34(51)                   |
| $\text{HNF} \tilde{X}^2A''$  | Comp. $r_e$  | 1.0288              | 1.3632              | 100.42                       |
|                              | Exp. $r_0^2$ | 1.06 <sup>a</sup>   | 1.37                | 105 <sup>a</sup>             |
| $\text{NF}_2 \tilde{X}^2B_1$ | Comp. $r_e$  | 1.3394              |                     | 103.20                       |
|                              | Exp. $r_0^3$ | 1.3494              |                     | 103.33                       |
| $\text{CH}_3 \tilde{X}^2A_2$ | Comp. $r_e$  | 1.0768              |                     | 120.00                       |
|                              | Exp. $r_0^4$ | 1.079               |                     | 120.0                        |
| $\text{HOO} \tilde{X}^2A''$  | Comp. $r_e$  | 0.9692              | 1.3246              | 104.62                       |
|                              | Exp. $r_e^5$ | 0.9702              | 1.3311              | 104.48                       |
|                              | Exp. $r_0^6$ | 0.9707              | 1.3339              | 104.15                       |
| $\text{HOO} \tilde{A}^2A'$   | Comp. $r_e$  | 0.9670              | 1.3912              | 102.23                       |
|                              | Exp. $r_e^5$ | 0.9676              | 1.3986              | 101.81                       |
| $\text{HCO} \tilde{X}^2A'$   | Comp. $r_e$  | 1.1161              | 1.1729              | 125.02                       |
|                              | Exp. $r_e^7$ | 1.1191(50)          | 1.1754(15)          | 124.43(25)                   |
|                              | Exp. $r_0^8$ | 1.125(5)            | 1.175(1)            | 124.95(25)                   |
| Continued on next page       |              |                     |                     |                              |

**Table 1 – continued from previous page**

| Radical and state                 | Structure                | $r_{XY}/\text{\AA}$ | $r_{YZ}/\text{\AA}$ | $\theta_{XYZ}/\text{degree}$ |
|-----------------------------------|--------------------------|---------------------|---------------------|------------------------------|
| FCO $\tilde{X}^2A'$               |                          |                     |                     |                              |
|                                   | Comp. $r_e$              | 1.3210              | 1.1643              | 127.98                       |
|                                   | Exp. $r_e$ <sup>9</sup>  | 1.334(60)           | 1.169(60)           | 127.3(6)                     |
| NO <sub>2</sub> $\tilde{X}^2A_1$  |                          |                     |                     |                              |
|                                   | Comp. $r_e$              | 1.1842              |                     | 134.96                       |
|                                   | Exp. $r_e$ <sup>10</sup> | 1.19389(4)          |                     | 133.857(3)                   |
| PH <sub>2</sub> $\tilde{X}^2B_1$  |                          |                     |                     |                              |
|                                   | Comp. $r_e$              | 1.4159              |                     | 92.03                        |
|                                   | Exp. $r_0$ <sup>11</sup> | 1.4260(71)          |                     | 91.65(98)                    |
| PH <sub>2</sub> $\tilde{A}^2A_1$  |                          |                     |                     |                              |
|                                   | Comp. $r_e$              | 1.3911              |                     | 121.89                       |
|                                   | Exp. $r_0$ <sup>12</sup> | 1.389               |                     | 123.2                        |
| ClO <sub>2</sub> $\tilde{X}^2B_1$ |                          |                     |                     |                              |
|                                   | Comp. $r_e$              | 1.4627              |                     | 116.82                       |
|                                   | Exp. $r_e$ <sup>14</sup> | 1.4698              |                     | 117.41                       |
|                                   | Exp. $r_0$ <sup>13</sup> | 1.474               |                     | 117.7                        |
| HSiS $\tilde{X}^2A'$              |                          |                     |                     |                              |
|                                   | Comp. $r_e$              | 1.5032              | 1.9520              | 119.94                       |
|                                   | Exp. $r_0$ <sup>15</sup> | 1.5140(fixed)       | 1.9537(19)          | 118.8(17)                    |
| CH <sub>2</sub> $\tilde{X}^3B_1$  |                          |                     |                     |                              |
|                                   | Comp. $r_e$              | 1.0764              |                     | 133.66                       |
|                                   | Exp. $r_e$ <sup>16</sup> | 1.0766(14)          |                     | 134.037(45)                  |

<sup>a</sup> Presumably inaccurate data.

Table 2: Computed e-SR tensor elements (in MHz) for NH<sub>2</sub> X<sup>2</sup>B<sub>1</sub>

HF/aug-cc-pVDZ

-6709.69 0.00 0.00

0.00 -1056.03 0.00

0.00 0.00 3.10

CCSD/aug-cc-pVDZ

-8194.70 0.00 0.00

0.00 -1262.64 0.00

0.00 0.00 6.57

CCSDT/aug-cc-pVDZ

-8301.15 0.00 0.00

0.00 -1276.88 0.00

0.00 0.00 6.72

London orb. HF/aug-cc-pVDZ

-6638.97 0.00 0.00

0.00 -1025.20 0.00

0.00 0.00 16.52

London orb. CCSD/aug-cc-pVDZ

-8139.17 0.00 0.00

0.00 -1236.68 0.00

0.00 0.00 19.40

HF/aug-cc-pVTZ

-6887.94 0.00 0.00

0.00 -1097.98 0.00

0.00 0.00 3.18

CCSD/aug-cc-pVTZ

-8828.50 0.00 0.00

```
0.00 -1356.06 0.00
0.00 0.00 6.29
CCSDT/aug-cc-pVTZ
-8993.59 0.00 0.00
0.00 -1379.52 0.00
0.00 0.00 6.44
London orb. HF/aug-cc-pVTZ
-6815.61 0.00 0.00
0.00 -1058.09 0.00
0.00 0.00 15.95
London orb. CCSD/aug-cc-pVTZ
-8763.19 0.00 0.00
0.00 -1319.30 0.00
0.00 0.00 18.63
HF/aug-cc-pVQZ
-7002.16 0.00 0.00
0.00 -1115.21 0.00
0.00 0.00 2.89
CCSD/aug-cc-pVQZ
-9032.72 0.00 0.00
0.00 -1382.97 0.00
0.00 0.00 6.16
HF/aug-cc-pV5Z
-7052.95 0.00 0.00
0.00 -1123.64 0.00
0.00 0.00 2.84
CCSD/aug-cc-pV5Z
```

```
-9112.71 0.00 0.00
0.00 -1395.09 0.00
0.00 0.00 6.15
HF/aug-cc-pCVTZ
-6960.39 0.00 0.00
0.00 -1111.03 0.00
0.00 0.00 2.99
AE CCSD/aug-cc-pCVTZ
-9001.08 0.00 0.00
0.00 -1378.60 0.00
0.00 0.00 6.14
AE CCSD/aug-cc-pCVTZ//experimental r0
-9034.30 0.00 0.00
0.00 -1371.57 0.00
0.00 0.00 6.09
FC CCSD/aug-cc-pCVTZ
-8912.59 0.00 0.00
0.00 -1370.12
0.00 0.00 6.16
HF/cc-pCVTZ
-7011.56 0.00 0.00
0.00 -1081.66 0.00
0.00 0.00 -1.46
CCSD/cc-pCVTZ
-8922.05 0.00 0.00
0.00 -1327.27 0.00
0.00 0.00 2.20
```

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

-8868.76 0.00 0.00

0.00 -1308.98 0.00

0.00 0.00 74.40

Table 3: Computed e-SR tensor elements (in MHz) for NHF X2A"

HF/aug-cc-pVDZ

-6066.08 104.97 0.00

1500.74 -234.81 0.00

0.00 0.00 16.31

CCSD/aug-cc-pVDZ

-8158.81 114.83 0.00

1645.14 -277.22 0.00

0.00 0.00 12.10

CCSDT/aug-cc-pVDZ

-8535.25 113.86 0.00

1631.09 -278.45 0.00

0.00 0.00 11.32

London orb. HF/aug-cc-pVDZ

-6006.94 104.46 0.00

1440.41 -229.71 0.00

0.00 0.00 19.38

London orb. CCSD/aug-cc-pVDZ

-8111.85 114.51 0.00

1579.11 -272.88 0.00

0.00 0.00 14.98

HF/aug-cc-pVTZ

-6255.88 106.91 0.00

1498.50 -240.94 0.00

0.00 0.00 16.65

CCSD/aug-cc-pVTZ

-8753.37 123.37 0.00



1725.06 -295.70 0.00

0.00 0.00 12.24

London orb. HF/aug-cc-pVTZ

-6191.26 106.87 0.00

1491.10 -236.11 0.00

0.00 0.00 19.02

London orb. CCSD/aug-cc-pVTZ

-8695.53 123.45 0.00

1710.86 -290.75 0.00

0.00 0.00 14.62

HF/aug-cc-pVQZ

-6343.55 108.59 0.00

1512.15 -244.10 0.00

0.00 0.00 16.82

CCSD/aug-cc-pVQZ

-8905.24 125.96 0.00

1754.84 -299.98 0.00

0.00 0.00 12.22

HF/aug-cc-pCVTZ

-6293.67 107.60 0.00

1499.82 -241.99 0.00

0.00 0.00 16.66

AE CCSD/aug-cc-pCVTZ

-8862.41 125.36 0.00

1745.76 -298.52 0.00

0.00 0.00 12.35

FC CCSD/aug-cc-pCVTZ

-8806.89 124.31 0.00

1727.71 -297.51 0.00

0.00 0.00 12.27

HF/cc-pCVTZ

-6285.77 110.29 0.00

1663.69 -247.10 0.00

0.00 0.00 16.80

AE CCSD/cc-pCVTZ

-8789.83 128.03 0.00

1974.16 -305.48 0.00

0.00 0.00 12.48

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

-8726.33 -513.58 0.00

1748.55 -293.44 0.00

0.00 0.00 20.74

Table 4: Computed e-SR tensor elements (in MHz) for NF2 X2B1

HF/aug-cc-pVDZ

-833.11 0.00 0.00

0.00 -70.03 0.00

0.00 0.00 17.26

CCSD/aug-cc-pVDZ

-927.02 0.00 0.00

0.00 -87.40 0.00

0.00 0.00 10.88

CCSDT/aug-cc-pVDZ

-926.40 0.00 0.00

0.00 -88.69 0.00

0.00 0.00 10.17

London orb. HF/aug-cc-pVDZ

-815.87 0.00 0.00

0.00 -68.93 0.00

0.00 0.00 18.34

London orb. HF/aug-cc-pVDZ

-912.13 0.00 0.00

0.00 -86.38 0.00

0.00 0.00 11.99

HF/aug-cc-pVTZ

-849.23 0.00 0.00

0.00 -71.92 0.00

0.00 0.00 17.48

CCSD/aug-cc-pVTZ

-989.73 0.00 0.00

```
0.00 -93.62 0.00
0.00 0.00 11.04
London orb. HF/aug-cc-pVTZ
-836.71 0.00 0.00
0.00 -70.27 0.00
0.00 0.00 18.37
London orb. CCSD/aug-cc-pVTZ
-977.04 0.00 0.00
0.00 -92.05 0.00
0.00 0.00 11.99
HF/aug-cc-pVQZ
-860.30 0.00 0.00
0.00 -72.98 0.00
0.00 0.00 17.67
CCSD/aug-cc-pVQZ
-1007.25 0.00 0.00
0.00 -95.12 0.00
0.00 0.00 11.06
HF/aug-cc-pCVTZ
-852.57 0.00 0.00
0.00 -72.39 0.00
0.00 0.00 17.50
AE CCSD/aug-cc-pCVTZ
-1001.50 0.00 0.00
0.00 -94.66 0.00
0.00 0.00 11.14
AE CCSD/aug-cc-pCVTZ//experimental r0
```

-989.77 0.00 0.00

0.00 -93.83 0.00

0.00 0.00 11.42

FC CCSD/aug-cc-pCVTZ

-995.31 0.00 0.00

0.00 -94.35 0.00

0.00 0.00 11.06

HF/cc-pCVTZ

-872.99 0.00 0.00

0.00 -74.06 0.00

0.00 0.00 17.48

AE CCSD/cc-pCVTZ

-1031.97 0.00 0.00

0.00 -98.03 0.00

0.00 0.00 11.28

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

-983.97 0.00 0.00

0.00 -92.67 0.00

0.00 0.00 13.69

Table 5: Computed e-SR tensor elements (in MHz) for CH<sub>3</sub> X2A<sub>2</sub>

HF/aug-cc-pVDZ

-305.59 0.00 0.00

0.00 -305.59 0.00

0.00 0.00 2.64

CCSD/aug-cc-pVDZ

-365.19 0.00 0.00

0.00 -365.19 0.00

0.00 0.00 3.93

CCSDT/aug-cc-pVDZ

-369.51 0.00 0.00

0.00 -369.51 0.00

0.00 0.00 3.99

London orb. HF/aug-cc-pVDZ

-286.37 0.00 0.00

0.00 -286.37 0.00

0.00 0.00 7.55

London orb. CCSD/aug-cc-pVDZ

-347.98 0.00 0.00

0.00 -347.98 0.00

0.00 0.00 8.74

HF/aug-cc-pVTZ

-319.82 0.00 0.00

0.00 -319.82 0.00

0.00 0.00 2.11

CCSD/aug-cc-pVTZ

-391.55 0.00 0.00

```
0.00 -391.55 0.00
0.00 0.00 3.53
CCSDT/aug-cc-pVTZ
-398.48 0.00 0.00
0.00 -398.48 0.00
0.00 0.00 3.60
London orb. HF/aug-cc-pVTZ
-297.30 0.00 0.00
0.00 -297.30 0.00
0.00 0.00 7.86
London orb. CCSD/aug-cc-pVTZ
-370.57 0.00 0.00
0.00 -370.57 0.00
0.00 0.00 9.07
HF/aug-cc-pVQZ
-325.54 0.00 0.00
0.00 -325.54 0.00
0.00 0.00 1.86
CCSD/aug-cc-pVQZ
-399.38 0.00 0.00
0.00 -399.38 0.00
0.00 0.00 3.37
London orb. HF/aug-cc-pVQZ
-301.83 0.00 0.00
0.00 -301.83 0.00
0.00 0.00 7.70
London orb. CCSD/aug-cc-pVQZ
```

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-376.90 0.00 0.00
0.00 -376.90 0.00
0.00 0.00 8.98
HF/aug-cc-pV5Z
-328.02 0.00 0.00
0.00 -328.02 0.00
0.00 0.00 1.84
CCSD/aug-cc-pV5Z
-402.79 0.00 0.00
0.00 -402.79 0.00
0.00 0.00 3.36
HF/aug-cc-pCVTZ
-324.65 0.00 0.00
0.00 -324.65 0.00
0.00 0.00 1.93
AE CCSD/aug-cc-pCVTZ
-399.41 0.00 0.00
0.00 -399.41 0.00
0.00 0.00 3.42
AE CCSD/aug-cc-pCVTZ//experimental r0
-397.11 0.00 0.00
0.00 -397.11 0.00
0.00 0.00 3.39
FC CCSD/aug-cc-pCVTZ
-396.51 0.00 0.00
0.00 -396.51 0.00
0.00 0.00 3.42
```



HF/cc-pCVTZ

-321.37 0.00 0.00

0.00 -321.37 0.00

0.00 0.00 1.23

AE CCSD/cc-pCVTZ

-391.86 0.00 0.00

0.00 -391.86 0.00

0.00 0.00 2.82

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

-373.56 0.00 0.00

0.00 -373.56 0.00

0.00 0.00 24.48

Table 6: Computed e-SR tensor elements (in MHz) for H<sub>2</sub>O X<sup>2</sup>A''

HF/aug-cc-pVDZ

-51623.81 -81.66 0.00

-294.99 -413.60 0.00

0.00 0.00 3.54

CCSD/aug-cc-pVDZ

-46298.60 -137.93 0.00

-1157.66 -410.29 0.00

0.00 0.00 5.40

CCSDT/aug-cc-pVDZ

-45286.27 -140.05 0.00

-1210.26 -399.06 0.00

0.00 0.00 6.21

London orb. HF/aug-cc-pVDZ

-51552.13 -81.58 0.00

-287.14 -407.59 0.00

0.00 0.00 6.86

London orb. CCSD/aug-cc-pVDZ

-46248.89 -137.80 0.00

-1127.03 -405.61 0.00

0.00 0.00 8.77

HF/aug-cc-pVTZ

-49298.96 -72.33 0.00

-244.60 -417.06 0.00

0.00 0.00 3.99

CCSD/aug-cc-pVTZ

-50574.97 -144.65 0.00

```
-1236.32 -436.38 0.00
0.00 0.00 5.63
London orb. HF/aug-cc-pVTZ
-49213.02 -72.33 0.00
-243.23 -411.22 0.00
0.00 0.00 6.97
London orb. CCSD/aug-cc-pVTZ
-50496.09 -144.87 0.00
-1211.61 -430.82 0.00
0.00 0.00 8.64
HF/aug-cc-pVQZ
-49769.15 -72.57 0.00
-235.04 -423.37 0.00
0.00 0.00 3.97
CCSD/aug-cc-pVQZ
-51745.30 -147.11 0.00
-1253.38 -445.94 0.00
0.00 0.00 5.75
HF/aug-cc-pCVTZ
-49566.99 -72.63 0.00
-239.55 -420.75 0.00
0.00 0.00 4.06
AE CCSD/aug-cc-pCVTZ
-51616.62 -146.54 0.00
-1237.39 -443.01 0.00
0.00 0.00 5.63
AE CCSD/aug-cc-pCVTZ//experimental re
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```
-52276.70 -145.77 0.00
-1254.33 -443.38 0.00
0.00 0.00 5.52
AE CCSD/aug-cc-pCVTZ//experimental r0
-52433.75 -142.74 0.00
-1219.42 -444.05 0.00
0.00 0.00 5.47
FC CCSD/aug-cc-pCVTZ
-51021.91 -145.86 0.00
-1239.80 -440.55 0.00
0.00 0.00 5.64
HF/cc-pCVTZ
-50621.55 -75.34 0.00
-418.24 -427.36 0.00
0.00 0.00 4.94
AE CCSD/cc-pCVTZ
-52192.80 -149.20 0.00
-1595.94 -447.76 0.00
0.00 0.00 5.75
from ESR tensor (AE CCSD/aug-cc-pCVTZ)
-51386.85 -2796.60 0.00
-1238.12 -434.01 0.00
0.00 0.00 19.28
```

Table 7: Computed e-SR tensor elements (in MHz) for H<sub>2</sub>O A<sub>2</sub>A'

HF/aug-cc-pVDZ

265815.03 39.04 0.00

-4473.17 10.38 0.00

0.00 0.00 -233.59

CCSD/aug-cc-pVDZ

73330.56 118.57 0.00

645.84 9.29 0.00

0.00 0.00 -291.78

CCSDT/aug-cc-pVDZ

53735.43 124.55 0.00

1031.71 9.73 0.00

0.00 0.00 -294.52

London orb. HF/aug-cc-pVDZ

265933.09 38.94 0.00

-4428.59 13.18 0.00

0.00 0.00 -228.42

London orb. CCSD/aug-cc-pVDZ

73448.73 118.16 0.00

671.99 12.95 0.00

0.00 0.00 -288.00

HF/aug-cc-pVTZ

884001.28 43.13 0.00

-15805.04 10.73 0.00

0.00 0.00 -236.09

CCSD/aug-cc-pVTZ

146622.18 122.11 0.00

```
-188.62 9.49 0.00
0.00 0.00 -308.68
London orb. HF/aug-cc-pVTZ
884121.41 43.08 0.00
-15821.87 13.24 0.00
0.00 0.00 -231.35
London orb. CCSD/aug-cc-pVTZ
146738.78 121.34 0.00
-152.12 12.86 0.00
0.00 0.00 -304.41
HF/aug-cc-pVQZ
1109996.51 44.14 0.00
-20037.96 10.84 0.00
0.00 0.00 -239.74
CCSD/aug-cc-pVQZ
189778.58 128.49 0.00
-748.13 9.46 0.00
0.00 0.00 -315.22
HF/aug-cc-pCVTZ
926758.96 42.26 0.00
-16602.97 10.86 0.00
0.00 0.00 -238.41
AE CCSD/aug-cc-pCVTZ
161712.12 123.33 0.00
-434.78 9.41 0.00
0.00 0.00 -312.91
AE CCSD/aug-cc-pCVTZ//experimental re
```

223155.36 109.78 0.00

-1467.08 9.45 0.00

0.00 0.00 -310.83

FC CCSD/aug-cc-pCVTZ

154409.82 124.39 0.00

-267.90 9.42 0.00

0.00 0.00 -311.46

HF/cc-pCVTZ

670577.78 18.10 0.00

-12003.99 11.75 0.00

0.00 0.00 -241.42

AE CCSD/cc-pCVTZ

124193.19 121.87 0.00

279.81 9.68 0.00

0.00 0.00 -312.76

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

161909.56 8215.80 0.00

-435.28 20.86 0.00

0.00 0.00 -306.74

Table 8: Computed e-SR tensor elements (in MHz) for HCO X2A'

HF/aug-cc-pVDZ

10713.07 113.02 0.00

2080.49 54.67 0.00

0.00 0.00 -178.76

CCSD/aug-cc-pVDZ

11359.80 110.55 0.00

1420.38 30.94 0.00

0.00 0.00 -181.78

CCSDT/aug-cc-pVDZ

11367.46 112.14 0.00

1308.73 27.34 0.00

0.00 0.00 -180.95

London orb. HF/aug-cc-pVDZ

10778.22 113.74 0.00

2018.95 57.64 0.00

0.00 0.00 -175.49

London orb. CCSD/aug-cc-pVDZ

11440.74 111.28 0.00

1360.03 33.74 0.00

0.00 0.00 -179.20

HF/aug-cc-pVTZ

10983.71 116.62 0.00

2091.80 57.43 0.00

0.00 0.00 -186.29

CCSD/aug-cc-pVTZ

12126.51 118.73 0.00



```
1524.79 31.74 0.00
0.00 0.00 -198.07
CCSDT/aug-cc-pVTZ
12173.27 120.26 0.00
1387.66 28.20 0.00
0.00 0.00 -196.87
London orb. HF/aug-cc-pVTZ
11054.48 117.31 0.00
2092.49 60.83 0.00
0.00 0.00 -181.14
London orb. CCSD/aug-cc-pVTZ
12209.79 119.07 0.00
1509.10 35.81 0.00
0.00 0.00 -193.17
HF/aug-cc-pVQZ
11110.35 117.55 0.00
2106.13 57.66 0.00
0.00 0.00 -189.12
CCSD/aug-cc-pVQZ
12323.07 120.13 0.00
1548.37 31.41 0.00
0.00 0.00 -201.48
HF/aug-cc-pCVTZ
11010.43 116.70 0.00
2086.18 57.07 0.00
0.00 0.00 -187.68
AE CCSD/aug-cc-pCVTZ
```

```
12311.78 119.65 0.00
1538.05 31.47 0.00
0.00 0.00 -201.02
AE CCSD/aug-cc-pCVTZ//experimental r0
12018.34 119.52 0.00
1511.24 31.47 0.00
0.00 0.00 -199.90
AE CCSD/aug-cc-pCVTZ//experimental re
11933.85 118.43 0.00
1542.07 31.74 0.00
0.00 0.00 -202.01
FC CCSD/aug-cc-pCVTZ
12187.94 119.10 0.00
1524.19 31.40 0.00
0.00 0.00 -199.84
HF/cc-pCVTZ
11094.89 117.77 0.00
2209.80 57.60 0.00
0.00 0.00 -187.19
AE CCSD/cc-pCVTZ
12380.56 121.64 0.00
1745.15 33.08 0.00
0.00 0.00 -199.55
from ESR tensor (AE CCSD/aug-cc-pCVTZ)
12465.85 757.30 0.00
1529.22 41.74 0.00
0.00 0.00 -195.29
```

Table 9: Computed e-SR tensor elements (in MHz) for FCO X2A'

1816.8 -19.8 0

-19.8 13.849 0

0 0 -32.741

HF/aug-cc-pVDZ

2039.14 -17.22 0.00

73.11 22.31 0.00

0.00 0.00 -26.62

CCSD/aug-cc-pVDZ

1855.30 -19.96 0.00

-54.29 17.79 0.00

0.00 0.00 -26.51

CCSDT/aug-cc-pVDZ

1824.53 -19.75 0.00

-73.04 17.50 0.00

0.00 0.00 -26.35

London orb. HF/aug-cc-pVDZ

2054.35 -17.27 0.00

66.40 24.04 0.00

0.00 0.00 -24.14

London orb. CCSD/aug-cc-pVDZ

1881.48 -21.08 0.00

-64.72 19.84 0.00

0.00 0.00 -23.75

HF/aug-cc-pVTZ

2092.74 -17.52 0.00

73.94 23.53 0.00

```
0.00 0.00 -26.52
CCSD/aug-cc-pVTZ
1972.93 -21.16 0.00
-46.41 18.96 0.00
0.00 0.00 -28.41
London orb. HF/aug-cc-pVTZ
2118.02 -17.56 0.00
71.67 24.64 0.00
0.00 0.00 -24.66
London orb. CCSD/aug-cc-pVTZ
1979.03 -19.36 0.00
-34.70 20.64 0.00
0.00 0.00 -26.45
HF/aug-cc-pVQZ
2114.18 -17.85 0.00
74.60 23.92 0.00
0.00 0.00 -26.84
CCSD/aug-cc-pVQZ
1999.48 -21.55 0.00
-43.99 19.17 0.00
0.00 0.00 -28.82
HF/aug-cc-pCVTZ
2093.21 -17.62 0.00
74.23 23.78 0.00
0.00 0.00 -26.56
AE CCSD/aug-cc-pCVTZ
1995.16 -21.56 0.00
```

```
-46.11 19.23 0.00
0.00 0.00 -28.92
AE CCSD/aug-cc-pCVTZ//experimental re
1905.01 -21.55 0.00
-35.13 18.84 0.00
0.00 0.00 -30.34
FC CCSD/aug-cc-pCVTZ
1977.83 -21.33 0.00
-45.14 19.22 0.00
0.00 0.00 -28.55
HF/cc-pCVTZ
2108.27 -18.12 0.00
77.44 23.56 0.00
0.00 0.00 -27.91
AE CCSD/cc-pCVTZ
2028.74 -22.41 0.00
-42.75 18.82 0.00
0.00 0.00 -31.27
from ESR tensor (AE CCSD/aug-cc-pCVTZ)
2042.12 122.05 0.00
-45.27 21.92 0.00
0.00 0.00 -27.79
```

Table 10: Computed e-SR tensor elements (in MHz) for NO2 X2A1

HF/aug-cc-pVDZ

6034.32 0.00 0.00

0.00 24.55 0.00

0.00 0.00 -99.51

CCSD/aug-cc-pVDZ

5447.20 0.00 0.00

0.00 14.18 0.00

0.00 0.00 -81.28

CCSDT/aug-cc-pVDZ

5349.12 0.00 0.00

0.00 14.16 0.00

0.00 0.00 -78.21

London orb. HF/aug-cc-pVDZ

6058.98 0.00 0.00

0.00 25.59 0.00

0.00 0.00 -97.06

London orb. CCSD/aug-cc-pVDZ

5483.31 0.00 0.00

0.00 15.73 0.00

0.00 0.00 -79.90

HF/aug-cc-pVTZ

6202.00 0.00 0.00

0.00 24.74 0.00

0.00 0.00 -102.27

CCSD/aug-cc-pVTZ

5876.15 0.00 0.00

```
0.00 14.49 0.00
0.00 0.00 -88.57
London orb. HF/aug-cc-pVTZ
6240.34 0.00 0.00
0.00 25.99 0.00
0.00 0.00 -100.01
London orb. CCSD/aug-cc-pVTZ
5915.49 0.00 0.00
0.00 15.82 0.00
0.00 0.00 -84.11
HF/aug-cc-pVQZ
6264.28 0.00 0.00
0.00 24.81 0.00
0.00 0.00 -103.74
CCSD/aug-cc-pVQZ
5982.01 0.00 0.00
0.00 14.46 0.00
0.00 0.00 -90.46
HF/aug-cc-pCVTZ
6203.24 0.00 0.00
0.00 24.69 0.00
0.00 0.00 -103.05
AE CCSD/aug-cc-pCVTZ
5958.89 0.00 0.00
0.00 14.47 0.00
0.00 0.00 -90.22
AE CCSD/aug-cc-pCVTZ//experimental re
```

5580.15 0.00 0.00

0.00 14.37 0.00

0.00 0.00 -95.72

FC CCSD/aug-cc-pCVTZ

5900.01 0.00 0.00

0.00 14.50 0.00

0.00 0.00 -89.49

HF/cc-pCVTZ

6269.91 0.00 0.00

0.00 25.27 0.00

0.00 0.00 -108.24

AE CCSD/cc-pCVTZ

6108.90 0.00 0.00

0.00 14.38 0.00

0.00 0.00 -97.64

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

6042.58 0.00 0.00

0.00 19.01 0.00

0.00 0.00 -87.94



Table 11: Computed e-SR tensor elements (in MHz) for PH<sub>2</sub> X2B1

HF/aug-cc-pVDZ

-6571.94 0.00 0.00

0.00 -1683.66 0.00

0.00 0.00 -40.01

CCSD/aug-cc-pVDZ

-7688.23 0.00 0.00

0.00 -2190.56 0.00

0.00 0.00 -19.51

CCSDT/aug-cc-pVDZ

-7783.99 0.00 0.00

0.00 -2258.07 0.00

0.00 0.00 -17.84

London orb. HF/aug-cc-pVDZ

-6574.03 0.00 0.00

0.00 -1682.25 0.00

0.00 0.00 -33.51

London orb. CCSD/aug-cc-pVDZ

-7765.88 0.00 0.00

0.00 -2225.78 0.00

0.00 0.00 -14.08

HF/aug-cc-pVTZ

-6858.19 0.00 0.00

0.00 -1683.93 0.00

0.00 0.00 -39.30

CCSD/aug-cc-pVTZ

-8365.30 0.00 0.00

```
0.00 -2328.15 0.00
0.00 0.00 -18.85
CCSDT/aug-cc-pVTZ
-8534.56 0.00 0.00
0.00 -2427.61 0.00
0.00 0.00 -17.53
London orb. HF/aug-cc-pVTZ
-6850.86 0.00 0.00
0.00 -1677.21 0.00
0.00 0.00 -36.38
London orb. CCSD/aug-cc-pVTZ
-8372.36 0.00 0.00
0.00 -2327.75 0.00
0.00 0.00 -16.09
HF/aug-cc-pVQZ
-6611.37 0.00 0.00
0.00 -1631.49 0.00
0.00 0.00 -39.41
CCSD/aug-cc-pVQZ
-8145.48 0.00 0.00
0.00 -2269.84 0.00
0.00 0.00 -18.26
HF/aug-cc-pV5Z
-7115.85 0.00 0.00
0.00 -1718.49 0.00
0.00 0.00 -43.57
CCSD/aug-cc-pV5Z
```

-8821.33 0.00 0.00

0.00 -2420.41 0.00

0.00 0.00 -20.66

HF/aug-cc-pCVTZ

-7063.37 0.00 0.00

0.00 -1710.24 0.00

0.00 0.00 -42.39

AE CCSD/aug-cc-pCVTZ

-8763.20 0.00 0.00

0.00 -2421.09 0.00

0.00 0.00 -20.47

AE CCSD/aug-cc-pCVTZ//experimental r0

-8629.84 0.00 0.00

0.00 -2427.98 0.00

0.00 0.00 -20.58

FC CCSD/aug-cc-pCVTZ

-8651.52 0.00 0.00

0.00 -2380.52 0.00

0.00 0.00 -19.99

HF/cc-pCVTZ

-7044.71 0.00 0.00

0.00 -1653.59 0.00

0.00 0.00 -47.25

AE CCSD/cc-pCVTZ

-8648.06 0.00 0.00

0.00 -2313.78 0.00

0.00 0.00 -28.19

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

-8773.12 0.00 0.00

0.00 -2431.04 0.00

0.00 0.00 -2.06

Table 12: Computed e-SR elements (in MHz) for PH<sub>2</sub> A2A1

HF/aug-cc-pVDZ

43163.46 0.00 0.00

0.00 426.40 0.00

0.00 0.00 -869.19

CCSD/aug-cc-pVDZ

30246.20 0.00 0.00

0.00 177.55 0.00

0.00 0.00 -1075.82

CCSDT/aug-cc-pVDZ

29684.11 0.00 0.00

0.00 145.65 0.00

0.00 0.00 -1104.84

London orb. HF/aug-cc-pVDZ

43176.55 0.00 0.00

0.00 431.15 0.00

0.00 0.00 -871.87

London orb. CCSD/aug-cc-pVDZ

30429.93 0.00 0.00

0.00 183.02 0.00

0.00 0.00 -1089.71

HF/aug-cc-pVTZ

50388.76 0.00 0.00

0.00 496.83 0.00

0.00 0.00 -878.84

CCSD/aug-cc-pVTZ

33575.02 0.00 0.00

```
0.00 209.33 0.00
0.00 0.00 -1148.59
CCSDT/aug-cc-pVTZ
32568.47 0.00 0.00
0.00 163.89 0.00
0.00 0.00 -1195.17
London orb. HF/aug-cc-pVTZ
50408.15 0.00 0.00
0.00 501.29 0.00
0.00 0.00 -881.43
London orb. CCSD/aug-cc-pVTZ
33530.83 0.00 0.00
0.00 213.47 0.00
0.00 0.00 -1136.86
HF/aug-cc-pVQZ
49032.44 0.00 0.00
0.00 492.91 0.00
0.00 0.00 -846.64
CCSD/aug-cc-pVQZ
32865.71 0.00 0.00
0.00 206.18 0.00
0.00 0.00 -1115.29
HF/aug-cc-pV5Z
53234.96 0.00 0.00
0.00 543.21 0.00
0.00 0.00 -898.49
CCSD/aug-cc-pV5Z
```

```
35732.40 0.00 0.00
0.00 224.84 0.00
0.00 0.00 -1191.81
HF/aug-cc-pCVTZ
52576.19 0.00 0.00
0.00 535.17
0.00 0.00 -893.91
AE CCSD/aug-cc-pCVTZ
35154.70 0.00 0.00
0.00 230.61 0.00
0.00 0.00 -1200.28
AE CCSD/aug-cc-pCVTZ//experimental r0
38092.27 0.00 0.00
0.00 220.82 0.00
0.00 0.00 -1186.28
FC CCSD/aug-cc-pCVTZ
34861.09 0.00 0.00
0.00 224.50 0.00
0.00 0.00 -1172.59
HF/cc-pCVTZ
51273.58 0.00 0.00
0.00 497.97 0.00
0.00 0.00 -889.15
AE CCSD/cc-pCVTZ
34594.93 0.00 0.00
0.00 212.71 0.00
0.00 0.00 -1177.53
```

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

35154.15 0.00 0.00

0.00 252.43 0.00

0.00 0.00 -1202.58



Table 13: Computed e-SR tensor elements (in MHz) for ClO<sub>2</sub> X2B1

HF/aug-cc-pVDZ

-379.45 0.00 0.00

0.00 10.01 0.00

0.00 0.00 -2582.31

CCSD/aug-cc-pVDZ

-251.67 0.00 0.00

0.00 7.20 0.00

0.00 0.00 -1690.91

CCSDT/aug-cc-pVDZ

-232.23 0.00 0.00

0.00 7.25 0.00

0.00 0.00 -1533.98

London orb. HF/aug-cc-pVDZ

-373.84 0.00 0.00

0.00 11.73 0.00

0.00 0.00 -2579.60

London orb. CCSD/aug-cc-pVDZ

-247.43 0.00 0.00

0.00 8.58 0.00

0.00 0.00 -1683.76

HF/aug-cc-pVTZ

-339.86 0.00 0.00

0.00 10.75 0.00

0.00 0.00 -2360.64

CCSD/aug-cc-pVTZ

-244.41 0.00 0.00

```
0.00 7.27 0.00
0.00 0.00 -1644.98
London orb. HF/aug-cc-pVTZ
-338.24 0.00 0.00
0.00 11.72 0.00
0.00 0.00 -2366.72
London orb. CCSD/aug-cc-pVTZ
-242.98 0.00 0.00
0.00 8.15 0.00
0.00 0.00 -1649.25
HF/aug-cc-pVQZ
-331.04 0.00 0.00
0.00 9.99 0.00
0.00 0.00 -2305.56
CCSD/aug-cc-pVQZ
-239.40 0.00 0.00
0.00 6.85 0.00
0.00 0.00 -1614.39
HF/aug-cc-pCVTZ
-326.18 0.00 0.00
0.00 9.77 0.00
0.00 0.00 -2282.42
AE CCSD/aug-cc-pCVTZ
-238.44 0.00 0.00
0.00 6.79 0.00
0.00 0.00 -1600.62
AE CCSD/aug-cc-pCVTZ//experimental re
```

```
-244.67 0.00 0.00
0.00 6.89 0.00
0.00 0.00 -1650.64
AE CCSD/aug-cc-pCVTZ//experimental r0
-248.40 0.00 0.00
0.00 6.95 0.00
0.00 0.00 -1680.37
FC CCSD/aug-cc-pCVTZ
-234.94 0.00 0.00
0.00 6.81 0.00
0.00 0.00 -1575.07
HF/cc-pCVTZ
-332.95 0.00 0.00
0.00 10.72 0.00
0.00 0.00 -2387.00
AE CCSD/cc-pCVTZ
-242.66 0.00 0.00
0.00 7.29 0.00
0.00 0.00 -1717.10
from ESR tensor (AE CCSD/aug-cc-pCVTZ)
-1588.31 0.00 0.00
0.00 -237.17 0.00
0.00 0.00 9.38
```

Table 14: Computed e-SR tensor elements (in MHz) for HSiS X2A'

HF/aug-cc-pVDZ

8662.65 135.87 0.00

5677.83 129.81 0.00

0.00 0.00 -163.38

CCSD/aug-cc-pVDZ

9847.63 146.19 0.00

5777.78 106.98 0.00

0.00 0.00 -207.64

CCSDT/aug-cc-pVDZ

10186.26 148.08 0.00

5619.60 101.36 0.00

0.00 0.00 -206.90

London orb. HF/aug-cc-pVDZ

8659.03 135.76 0.00

5399.30 125.09 0.00

0.00 0.00 -168.30

London orb. CCSD/aug-cc-pVDZ

9899.17 146.92 0.00

5589.76 103.21 0.00

0.00 0.00 -213.22

HF/aug-cc-pVTZ

8876.51 141.25 0.00

5658.50 133.61 0.00

0.00 0.00 -166.17

CCSD/aug-cc-pVTZ

10291.61 157.45 0.00

```
6090.95 113.44 0.00
0.00 0.00 -221.76
London orb. HF/aug-cc-pVTZ
8889.87 141.31 0.00
5611.40 132.62 0.00
0.00 0.00 -166.67
London orb. CCSD/aug-cc-pVTZ
10346.67 157.98 0.00
6179.88 111.86 0.00
0.00 0.00 -222.20
HF/aug-cc-pVQZ
8659.99 136.37 0.00
5670.76 133.77 0.00
0.00 0.00 -159.37
CCSD/aug-cc-pVQZ
9988.68 151.65 0.00
6077.81 112.85 0.00
0.00 0.00 -211.83
HF/aug-cc-pCVTZ
9157.28 145.07 0.00
5815.50 136.67 0.00
0.00 0.00 -169.26
AE CCSD/aug-cc-pCVTZ
10666.33 164.14 0.00
6374.12 116.79 0.00
0.00 0.00 -235.00
FC CCSD/aug-cc-pCVTZ
```

10595.00 161.90 0.00

6292.12 116.59 0.00

0.00 0.00 -227.36

HF/cc-pCVTZ

9194.72 145.83 0.00

5869.97 137.76 0.00

0.00 0.00 -171.47

AE CCSD/cc-pCVTZ

10675.77 164.63 0.00

6501.96 118.71 0.00

0.00 0.00 -237.36

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

10672.52 282.27 0.00

6353.62 116.83 0.00

0.00 0.00 -236.02

Table 15: Computed e-SR tensor elements (in MHz) for CH<sub>2</sub> X3B1

HF/aug-cc-pVDZ

25.37 0.00 0.00

0.00 -114.77 0.00

0.00 0.00 -83.30

CCSD/aug-cc-pVDZ

24.68 0.00 0.00

0.00 -139.15 0.00

0.00 0.00 -106.81

CCSDT/aug-cc-pVDZ

24.49 0.00 0.00

0.00 -140.66 0.00

0.00 0.00 -108.37

London orb. HF/aug-cc-pVDZ

151.96 0.00 0.00

0.00 -101.13 0.00

0.00 0.00 -71.95

London orb. CCSD/aug-cc-pVDZ

147.57 0.00 0.00

0.00 -126.42 0.00

0.00 0.00 -96.22

HF/aug-cc-pVTZ

20.28 0.00 0.00

0.00 -121.70 0.00

0.00 0.00 -88.80

CCSD/aug-cc-pVTZ

18.69 0.00 0.00

```
0.00 -151.89 0.00
0.00 0.00 -117.43
CCSDT/aug-cc-pVTZ
18.25 0.00 0.00
0.00 -154.48 0.00
0.00 0.00 -120.04
London orb. HF/aug-cc-pVTZ
152.18 0.00 0.00
0.00 -106.32 0.00
0.00 0.00 -76.22
London orb. CCSD/aug-cc-pVTZ
149.95 0.00 0.00
0.00 -137.10 0.00
0.00 0.00 -105.53
HF/aug-cc-pVQZ
18.77 0.00 0.00
0.00 -124.22 0.00
0.00 0.00 -90.82
CCSD/aug-cc-pVQZ
17.06 0.00 0.00
0.00 -155.49 0.00
0.00 0.00 -120.41
HF/aug-cc-pV5Z
18.31 0.00 0.00
0.00 -125.21 0.00
0.00 0.00 -91.62
CCSD/aug-cc-pV5Z
```



```
16.53 0.00 0.00
0.00 -156.94 0.00
0.00 0.00 -121.62
HF/aug-cc-pCVTZ
17.85 0.00 0.00
0.00 -123.75 0.00
0.00 0.00 -90.61
AE CCSD/aug-cc-pCVTZ
16.45 0.00 0.00
0.00 -155.02 0.00
0.00 0.00 -120.22
AE CCSD/aug-cc-pCVTZ//experimental re
17.95 0.00 0.00
0.00 -153.90 0.00
0.00 0.00 -119.92
FC CCSD/aug-cc-pCVTZ
16.65 0.00 0.00
0.00 -153.94 0.00
0.00 0.00 -119.29
HF/cc-pCVTZ
18.06 0.00 0.00
0.00 -122.64 0.00
0.00 0.00 -90.43
AE CCSD/cc-pCVTZ
19.21 0.00 0.00
0.00 -152.16 0.00
0.00 0.00 -118.62
```

from ESR tensor (AE CCSD/aug-cc-pCVTZ)

219.04 0.00 0.00

0.00 -119.42 0.00

0.00 0.00 -89.12

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