**Table.** MRPT/aug-cc-pVTZ vertical transition energies (eV) of diazomethane.

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| State | Active space  (a1,b1,b2,a2) | State-average  (A1,B1,B2,A2) | CASSCF | CASPT2  NOIPEA | CASPT2  IPEA | SC-NEVPT2 | PC-NEVPT2 | CASPT3  NOIPEA | CASPT3  IPEA |
| 1A2(,\*) | (4,3,2,0) | (1,0,0,1) | 3.271a | **2.921** | **3.125** | **3.134** | **3.088** | **3.041** | **3.093** |
| 1B1(,3s) | (5,3,2,0) | (1,1,0,0) | 4.588b | **5.299** | **5.495** | **5.571** | **5.630** | **5.450** | **5.480** |
| 1A1(p,\*) | (4,4,2,0) | (3,0,0,0) | 5.653c | **5.925** | **6.213** | **6.281** | **6.232** | **6.129** | **6.182** |
| 3A2(,\*) | (4,3,2,0) | (1,0,0,1) | 3.023a | **2.666** | **2.870** | **2.882** | **2.835** | **2.792** | **2.843** |
| 3A1(p,\*) | (4,3,2,0) | (2,0,0,0) | 4.268a | **3.882** | **4.100** | **4.116** | **4.068** | **4.011** | **4.064** |
| 3B1(,3s) | (5,3,2,0) | (1,1,0,0) | 4.453b | **5.151** | **5.343** | **5.415** | **5.475** | **5.296** | **5.327** |
| 3A1(,3p) | (4,4,2,0) | (3,0,0,0) | 6.338c | **6.757** | **6.999** | **7.017** | **7.006** | **6.912** | **6.956** |
| 1A"[F](,\*) | (a’:6,a":3) | (A’:1,A":1) | 0.717a | **0.520** | **0.691** | **0.684** | **0.658** | **0.616** | **0.664** |

a Reference (10e,9o) active space including valence , σCN, σNN and σ\*CN, σ\*NN orbitals. b Reference (10e,10o) active space including valence , σCN, σNN and σ\*CN, σ\*NN, Rydberg 3s orbitals. c Reference (10e,10o) active space including valence , σCN, σNN and σ\*CN, σ\*NN, Rydberg 3p orbitals.