

Supporting information for

Carbon Dioxide Activation by Discandium Dioxide Cation in the Gas Phase: A Combined Investigation of Infrared Photodissociation Spectroscopy and DFT Calculations

Pengcheng Liu,^{1,2,†} Jia Han,^{3,†} Yan Chen,⁴ Shun Lu,⁴ Quyan Su,⁴

Xiaoguo Zhou,^{3,4,*} Weijun Zhang^{1,*}

1. *Anhui Institute of Optics and Fine Mechanics, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, China*
2. *Science Island Branch, Graduate School, University of Science and Technology of China, Hefei 230026, China*
3. *Hefei National Laboratory for Physical Sciences at the Microscale, University of Science and Technology of China, Hefei 230026, China*
4. *Department of Chemical Physics, University of Science and Technology of China, Hefei 230026, China*

Contents:

Figure S1. Schematic view of the experimental instrument consisting of a collinear tandem time-of-flight mass spectrometer coupled with a pulsed laser vaporization ion source.

Figure S2. The energetically lowest-lying isomer structures of Sc_2O_2^+ cation in the doublet, quartet and sextet states calculated at the PBE0-D3(BJ)/def2-TZVP level of theory, respectively. Relative Gibbs free energies (at 298 K) are given in kcal/mol. Bond lengths are noted in Å.

Figure S3. NPA (Natural Population Analysis) atomic charge distribution of the lowest-energy isomers in N, S and D groups calculated at the PBE0-D3(BJ)/def2-TZVP level of theory.

Figure S4. Typical TOF mass spectrum produced by pulsed laser vaporization of a scandium metal target in expansion of helium seeded with carbon dioxide.

Table S1. Binding energies of successive CO₂ ligands calculated for isomers of [Sc₂O₂(CO₂)_n]⁺ (n = 1–4) complex ions at the PBE0-D3(BJ)/def2-TZVP level of theory.

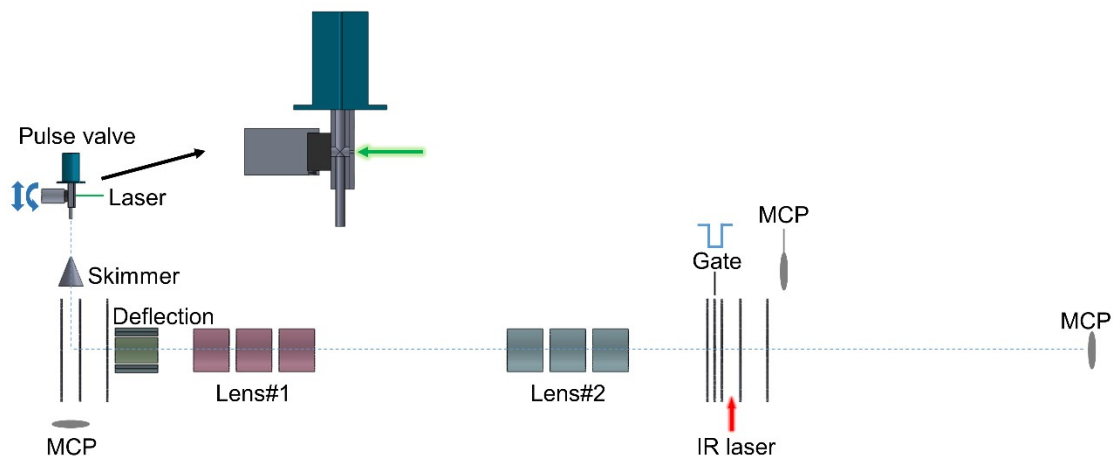


Figure S1. Schematic view of the experimental instrument consisting of a collinear tandem time-of-flight mass spectrometer coupled with a pulsed laser vaporization ion source.

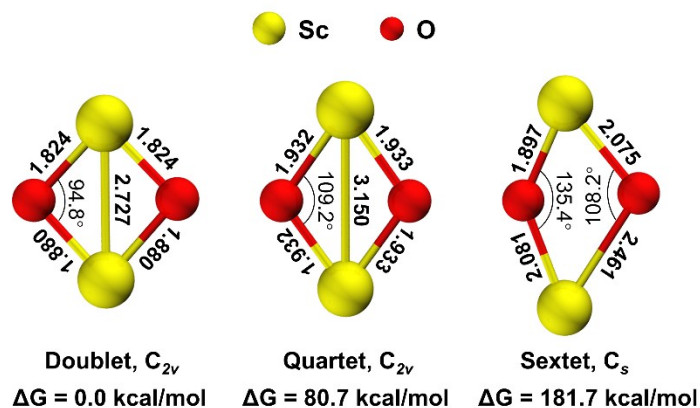


Figure S2. The energetically lowest-lying isomer structures of Sc₂O₂⁺ cation in the doublet, quartet and sextet states calculated at the PBE0-D3(BJ)/def2-TZVP level of theory, respectively. Relative Gibbs free energies (at 298 K) are given in kcal/mol. Bond lengths are noted in Å.

$$\text{Distortion energy}([\text{Sc}_2\text{O}_2]) = E([\text{Sc}_2\text{O}_2]_{\text{TS}}) - E([\text{Sc}_2\text{O}_2]_{\text{free}})$$

$$\text{Distortion energy}([\text{CO}_2]) = E([\text{CO}_2]_{\text{TS}}) - E([\text{CO}_2]_{\text{free}})$$

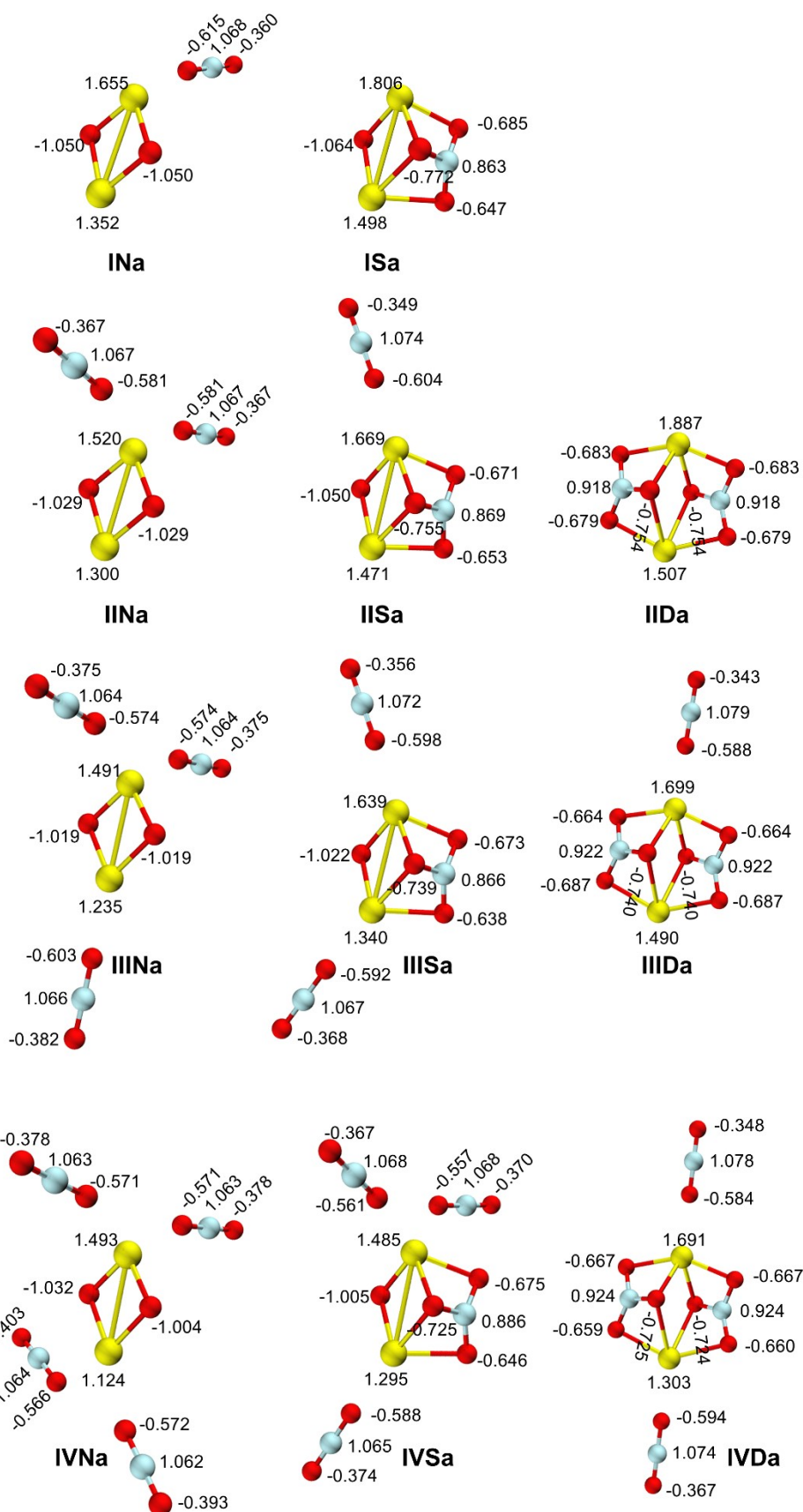


Figure S3. NPA (Natural Population Analysis) atomic charge distribution of the lowest-energy isomers in N, S and D groups calculated at the PBE0-D3(BJ)/def2-TZVP level of theory.

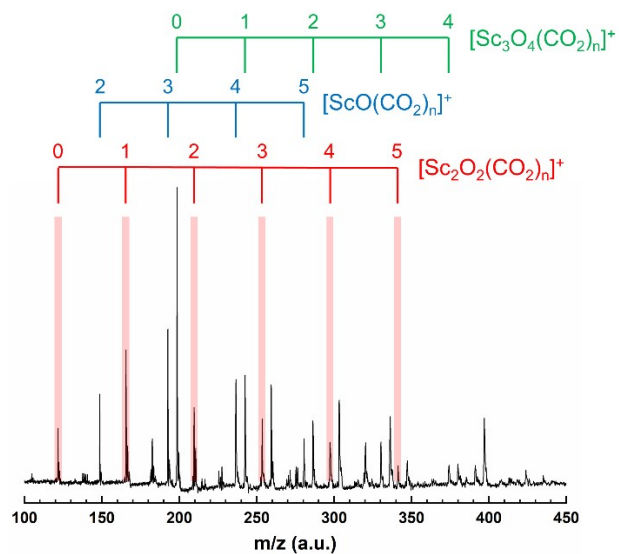


Figure S4. Typical TOF mass spectrum produced by pulsed laser vaporization of a scandium metal target in expansion of helium seeded with carbon dioxide.

Table S1. Binding energies of CO_2 ligands calculated for isomers of $[\text{Sc}_2\text{O}_2(\text{CO}_2)_n]^+$ ($n = 1-4$) complex ions at the PBE0-D3(BJ)/def2-TZVP level of theory.

	Binding energy (kcal/mol)			
	Na	Nb	Sa	Da
I	17.7			
II	17.4	15.0	20.8	
III	13.1	11.6	17.9	22.4
IV	11.1		15.9	17.2