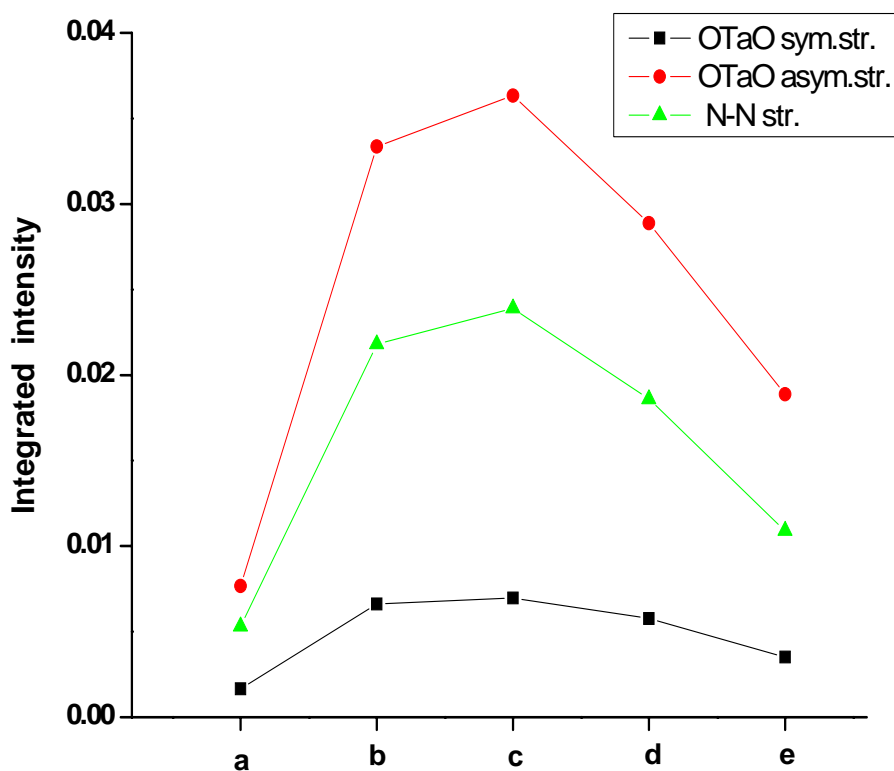


## Supporting Information

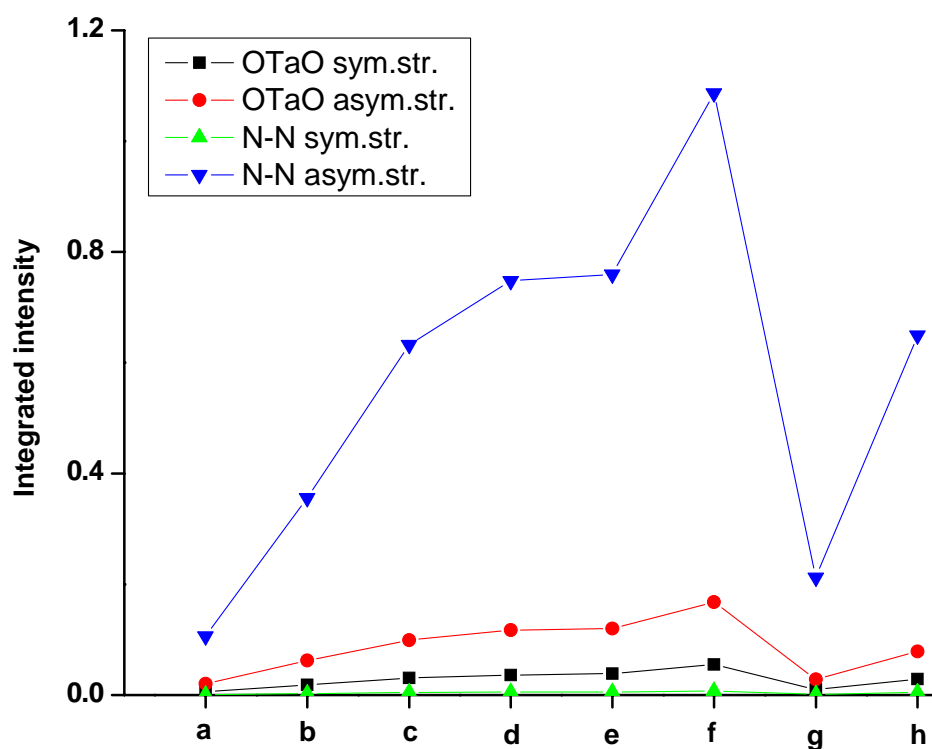
### Tantalum Dioxide Complexes with Dinitrogen. Formation and Characterization of the Side-on and End-on Bonded $\text{TaO}_2(\text{NN})_x$ ( $x=1-3$ ) Complexes

Caixia Wang, Jia Zhuang, Guanjun Wang, Mohua Chen, Yanying Zhao, Xuming Zheng,

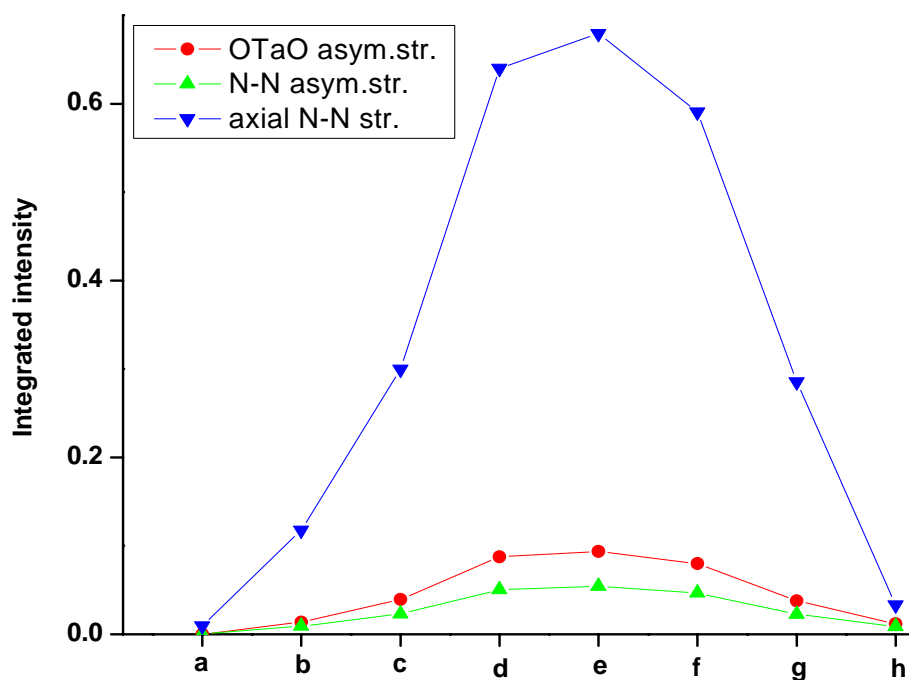
Mingfei Zhou



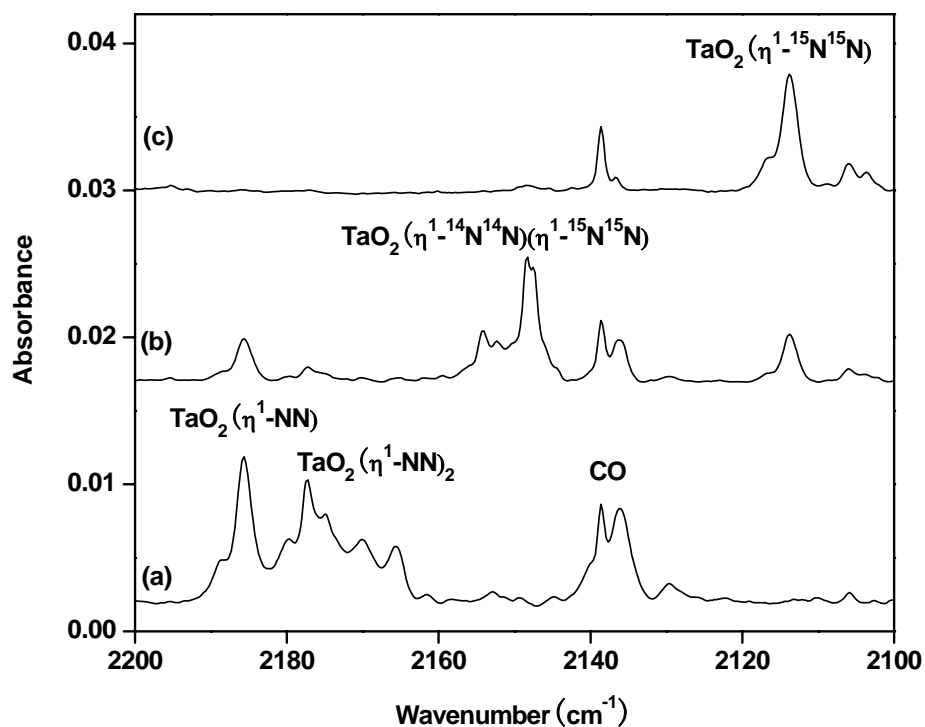
**Figure S1.** Plots of the integrated intensities of the observed absorptions assigned to  $\text{TaO}_2(\eta^1\text{-NN})$  as a function of annealing and photolysis. (a) 1 h of sample deposition at 4 K, (b) after 25 K annealing, (c) after 30 K annealing, (d) after 35 K annealing, and (e) after 40 K annealing.



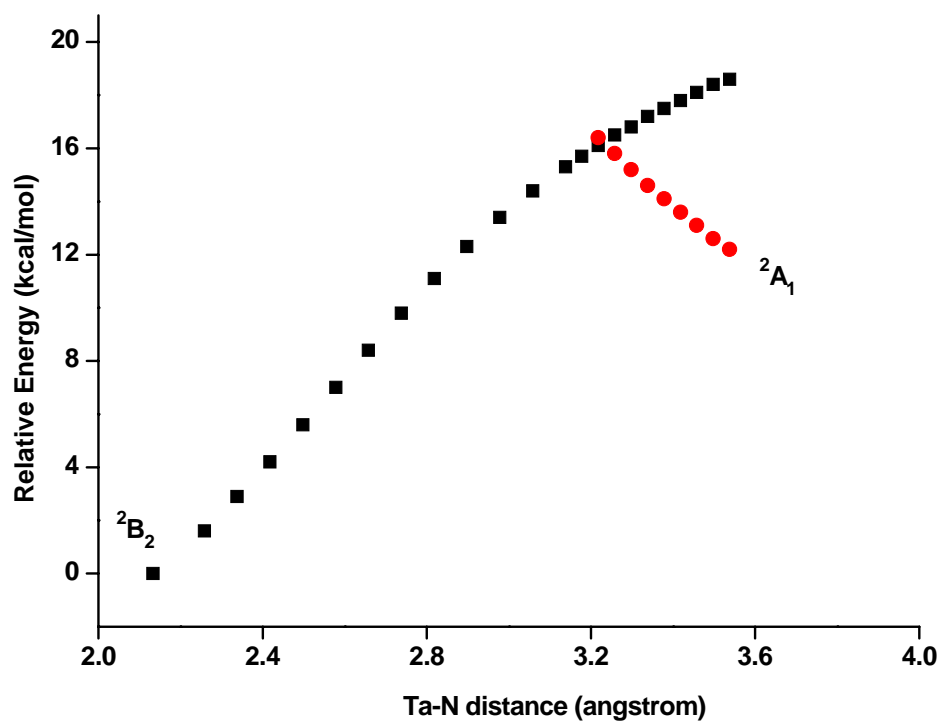
**Figure S2.** Plots of the integrated intensities of the observed absorptions assigned to  $\text{TaO}_2(\eta^1\text{-NN})_2$  as a function of annealing and photolysis. (a) 1 h of sample deposition at 4 K, (b) after 25 K annealing, (c) after 30 K annealing, (d) after 35 K annealing, (e) after 40 K annealing, (f) 30 min after 40 K annealing, (g) after 20 min of visible light irradiation ( $\lambda > 500$  nm), and (h) after 20 min of UV-visible light irradiation ( $300 < \lambda < 580$  nm).



**Figure S3.** Plots of the integrated intensities of the observed absorptions assigned to  $\text{TaO}_2(\eta^1\text{-NN})_3$  as a function of annealing and photolysis. (a) 1 h of sample deposition at 4 K, (b) after 25 K annealing, (c) after 30 K annealing, (d) after 35 K annealing, (e) after 40 K annealing, (f) 30 min after 40 K annealing, (g) after 20 min of visible light irradiation ( $\lambda > 500$  nm), and (h) after 20 min of UV-visible light irradiation ( $300 < \lambda < 580$  nm). (The symmetric OTaO stretching mode is overlapped by the antisymmetric OTaO stretching mode of  $\text{TaO}_2$ , and is not plotted).



**Figure S4.** Infrared spectra in the 2200-2100 cm<sup>-1</sup> region from codeposition of laser-evaporated tantalum oxides with isotopic-labeled dinitrogen in excess argon (Spectrum taken after 25 K annealing). (a) 0.1% <sup>14</sup>N<sub>2</sub>, (b) 0.1% <sup>14</sup>N<sub>2</sub> + 0.1% <sup>15</sup>N<sub>2</sub>, and (c) 0.1% <sup>15</sup>N<sub>2</sub>.



**Figure S5.** Potential energy curves along the axial Ta-N bond of  $^2B_2$  and  $^2A_1$  states

$TaO_2(\eta^1-NN)_3$  calculated at the MP2/6-311+G\*/SDD level of theory.