

Supplementary Information

## Visualizing rotational wave functions of electronically excited nitric oxide molecules by using ion imaging technique

Kenta Mizuse,<sup>\*a</sup> Nao Chizuwa,<sup>b</sup> Dai Ikeda,<sup>a</sup> Takashi Imajo,<sup>b</sup> and Yasuhiro Ohshima<sup>\*a,c</sup>

<sup>a</sup>Department of Chemistry, Tokyo Institute of Technology, 2-12-1-W4-9 Ookayama, Meguro, Tokyo 152-8550, Japan. E-mail: mizuse@chem.titech.ac.jp,

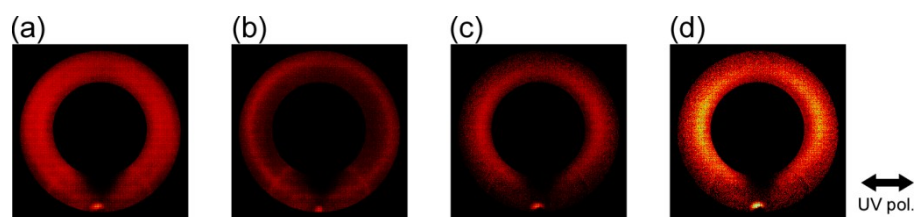
ohshima@chem.titech.ac.jp

<sup>b</sup>Department of Chemical and Biological Sciences, Japan Women's University, 2-8-1 Mejirodai, Bunkyo, Tokyo 112-8681, Japan.

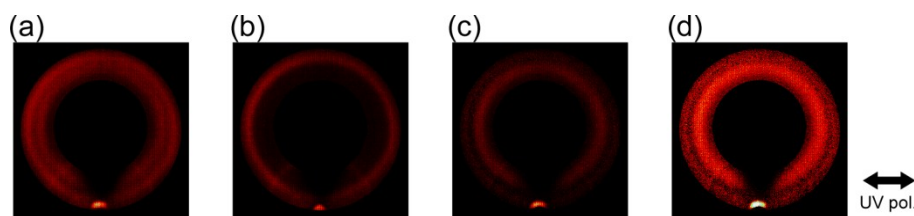
<sup>c</sup>Institute for Molecular Science, 38 Nishi-Gonaka, Myodaiji, Okazaki, Aichi 444-8585, Japan.

### Pump-probe delay dependence of the image

In the main text of this article, we have shown the images taken with the relatively long pump-probe delay of  $\sim 50$  ns. We also carried out the measurements with shorter,  $\sim 5$  ns delay (Fig. S1 and S2). Both Figs. S1d and S2d show similar trends to the Figs. 4d and 5d, respectively: Figure S1d shows anisotropic distribution along the UV pump polarization, while Fig. S2d shows an isotropic distribution. Note that the dark region always appears in the bottom of the images. This is due to the dead zone of the detector.



**Figure S1** Dissociative ionization imaging of NO with a  $44211\text{ cm}^{-1}$  UV pump ( $R_{12}(1/2)$  transition). (a)  $\text{N}^+$  ion image taken with a UV pump. (b) Probe-only  $\text{N}^+$  ion image. (c) Difference image between the pump + probe image (a) and the probe-only image (b). (d) Brightness-amplified image of (c). Only  $\text{N}^+$  ions from  $\text{NO}^{2+}$  were highlighted. The prepared state is  $|N S J |M\rangle = |2\ 1/2\ 3/2\ 1/2\rangle$ .



**Figure S2** Dissociative ionization imaging of NO with a  $44199\text{ cm}^{-1}$  UV pump ( $Q_{1a}(1/2)$  transition). (a)  $\text{N}^+$  ion image taken with a UV pump. (b) Probe-only  $\text{N}^+$  ion image. (c) Difference image between the pump + probe image (a) and the probe-only image (b). (d) Brightness-amplified image of (c). Only  $\text{N}^+$  ions from  $\text{NO}^{2+}$  were highlighted. The prepared state is  $|N S J |M\rangle = |0\ 1/2\ 1/2\ 1/2\rangle$ .