

Supporting Information

Gold-Atom-Mediated Charge Transfer in Plasmonic

Picocavity-Enabled CO₂ Dissociation

Xiaoting Ren, Tingting Wang, Huiru Xue, Maoshan Hou, Huan Chen, Jinping Li, Zhengkun Fu,

Lei Yan,* Zhenglong Zhang*

School of physics and information technology, Shaanxi Normal University, Shaanxi, Xi'an 710119, China.

*Corresponding author. Email: yanlei@snnu.edu.cn; zlzhang@snnu.edu.cn

The supplementary information includes:

Figs. S1-S4

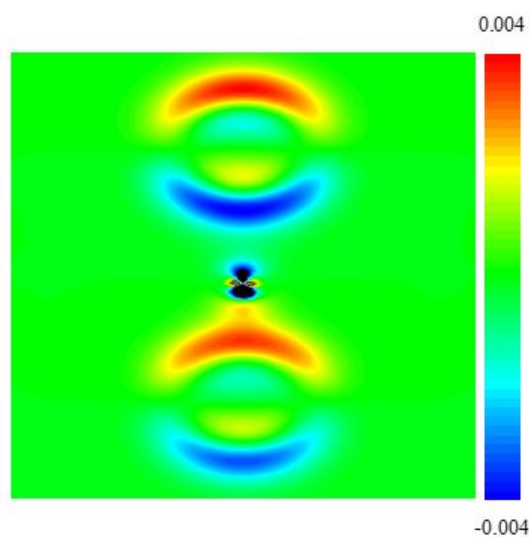


Fig. S1 Fourier transforms of induced charge distribution for the adatom-free system

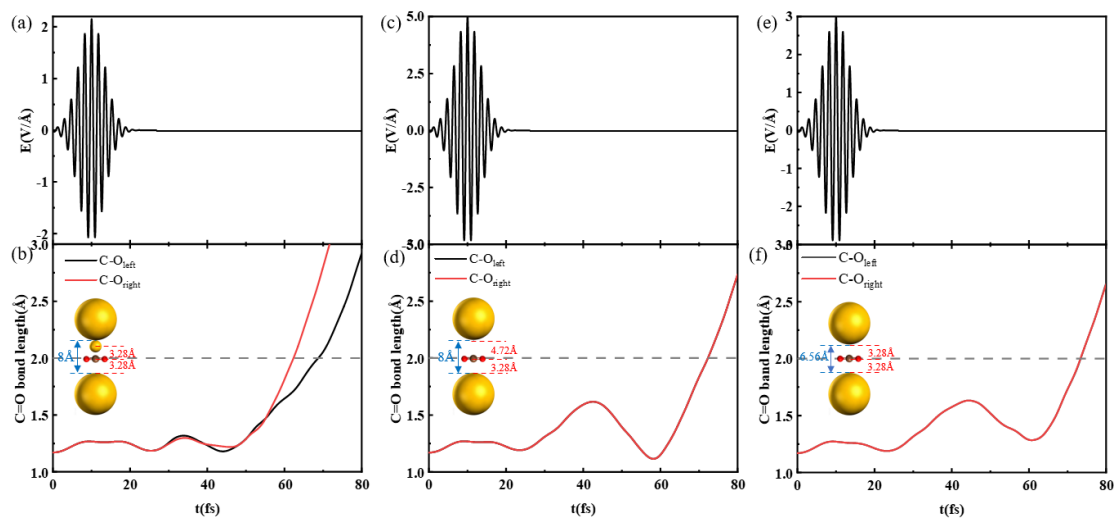


Fig. S2 Time evolved laser field with a maximum field strength of (a) 2.15 V/Å (c) 5.0 V/Å and (e) 3.0 V/Å ($\hbar\omega = 2.32$ eV) at the time $t = 10$ fs. Time evolution of the two C=O bond lengths of CO₂ for (b) the gold-adatom system (d) the adatom-free system and (f) the adatom-free-symmetry system

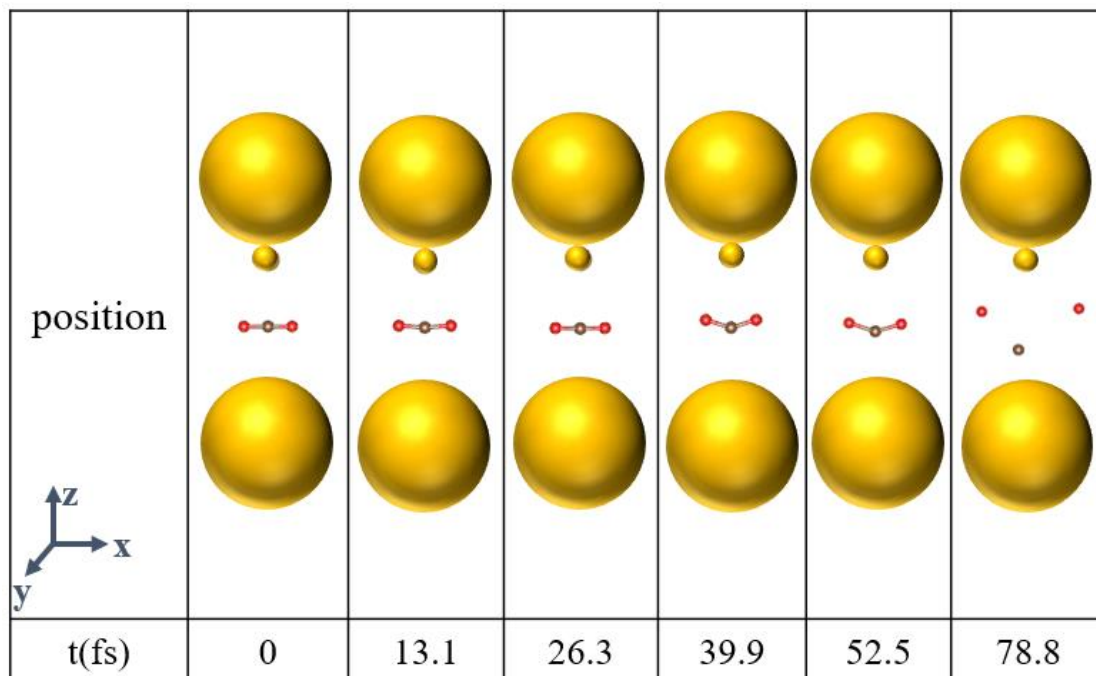


Fig. S3 Time evolution of the relative positions of the C and O atoms during CO₂ dissociation in the gold-adatom system (the metallic component is fixed)

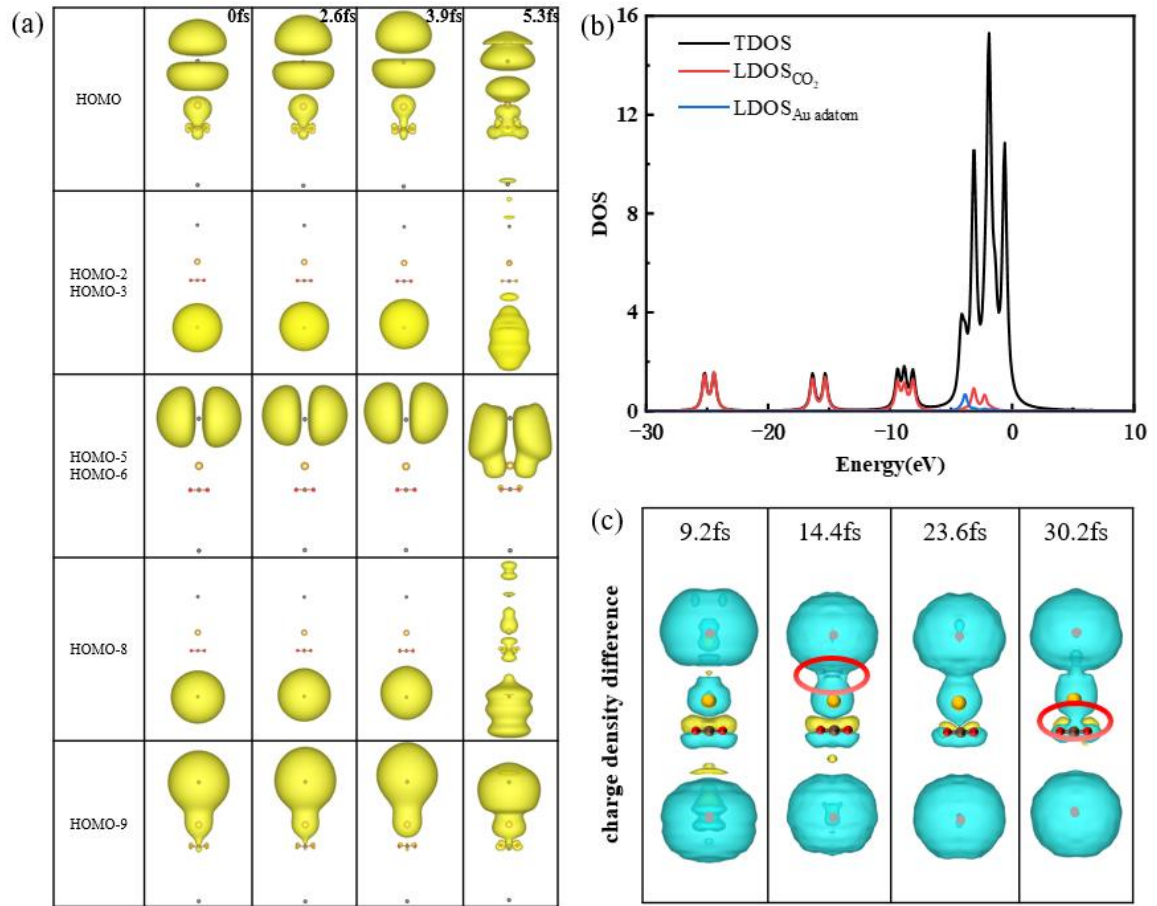


Fig. S4 (a) Time-resolved charge density at occupied orbitals in the gold-adatom system. The isosurface value is $0.004 \text{ \AA}^{-3}$. (b) The local density of states (LDOS) of CO₂ and the gold atom and total density of states (TDOS) in the gold-adatom system. (c) Time-resolved charge density difference in the gold-adatom system from 9.19 to 30.19 fs. The isosurface value is $0.005 \text{ \AA}^{-3}$. Red circles highlight two charge-transfer channels. Panels (a) and (c) are calculated under the laser with $E_{\text{max}} = 2.15 \text{ V/\AA}$ and $\hbar\omega = 2.32 \text{ eV}$.