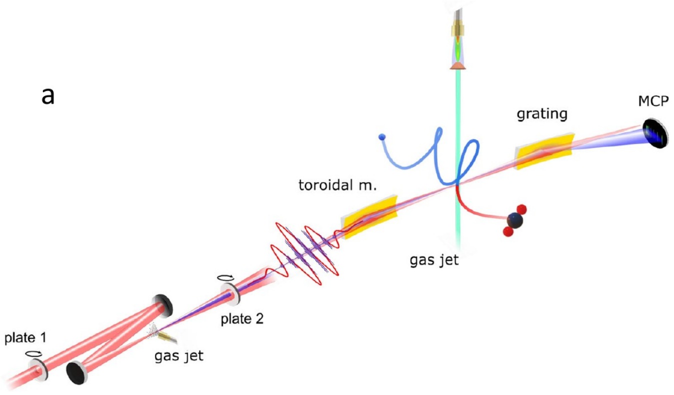
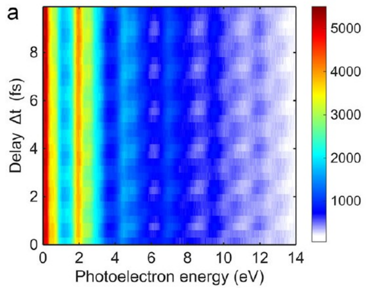
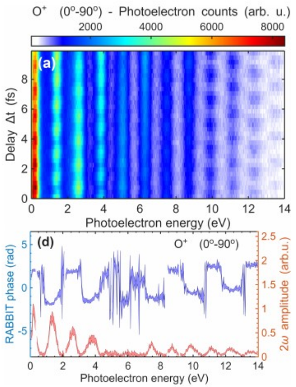




Attosecond coupling delay in ionization to entangled states of CO_2^+

Ioannis Makos, David Busto, Jakub Benda, Dominik Ertel,
Barbara Merzuk, Benjamin Steiner, Fabio Frassetto, Luca Poletto,
Claus Dieter Schröter, Thomas Pfeifer, Robert Moshammer,
Serguei Patchkovskii, Van-Hung Hoang, Uwe Thumm,
Zdeněk Mašín & Giuseppe Sansone



Deutsche Forschungsgemeinschaft
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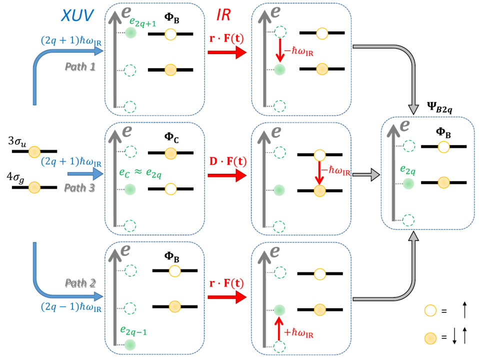
GAČR CZECH SCIENCE FOUNDATION



WavemiX 2026, June 2nd

Asymptotic RABBITT delays

RABBITT – measurement of XUV ionization delays
using XUV±IR two-photon ionization

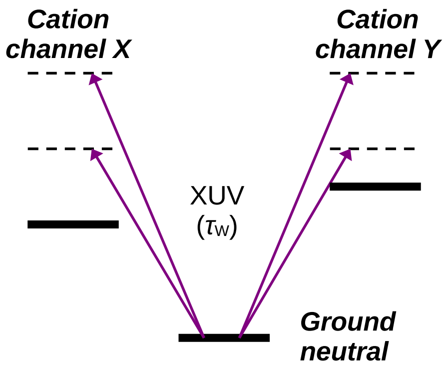
Asymptotic RABBITT delays

RABBITT – measurement of XUV ionization delays
using XUV±IR two-photon ionization

High-energy separability of RABBITT phases (delays)

$\tau_R \approx \tau_W$

τ_W – photoionization (Wigner) delay



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High-energy separability of RABBITT phases (delays)

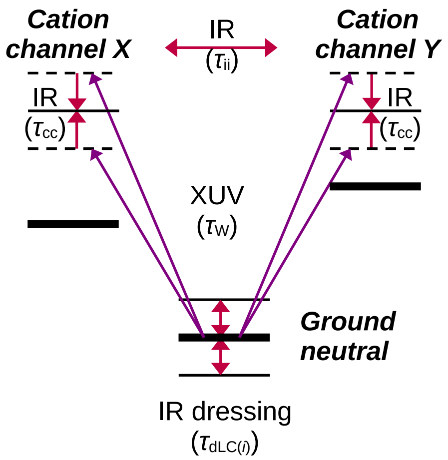
$$\tau_R \approx \tau_W + \tau_{cc} + \tau_{ii} + \tau_{dLC} + \ldots$$

τ_W – photoionization (Wigner) delay

τ_{cc} – continuum-continuum delay

τ_{ii} – ion-ion coupling delay

τ_{dLC} – dipole-laser coupling delay



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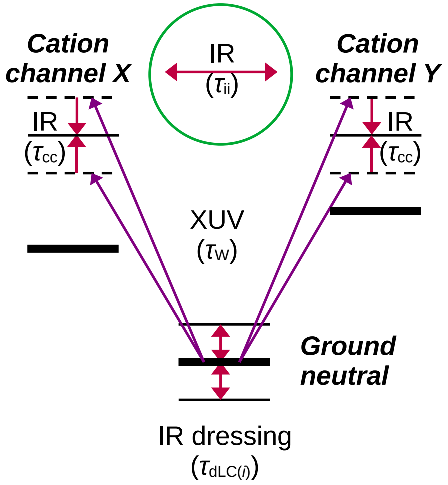

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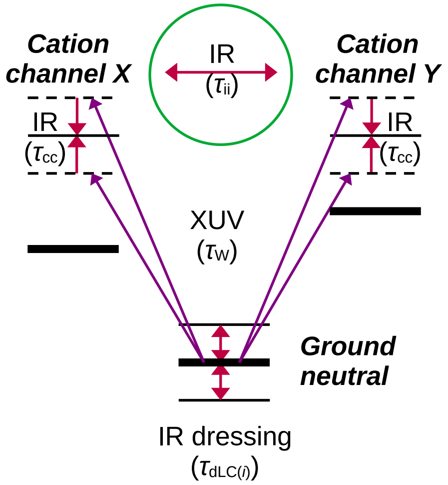
$$2\omega_{IR}\tau_R = \arg M_{XUV-IR} - \arg M_{XUV+IR}$$

$$M = \langle \Psi_f | D | \Psi_{i+\Omega} \rangle$$

final continuum state after XUV±IR ionization

dipole transition operator

intermediate continuum state after XUV ionization



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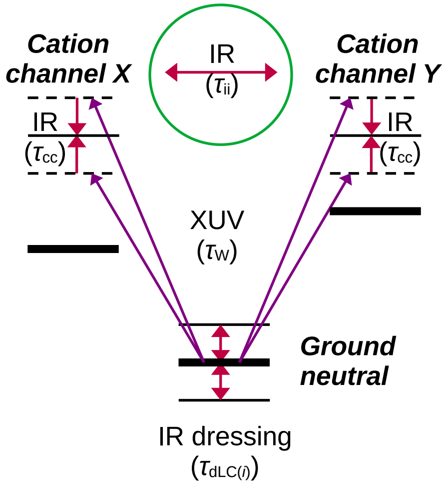

$$2\omega_{IR}\tau_R = \arg M_{XUV-IR} - \arg M_{XUV+IR}$$

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final continuum state after XUV±IR ionization

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intermediate continuum state after XUV ionization

$$D = \mathbf{r}_{pe} \cdot \boldsymbol{\epsilon}_{IR} + \mathbf{D}_{ion} \cdot \boldsymbol{\epsilon}_{IR}$$

$$M = M_{pe} + \delta M_{ion}$$

$$\tau_R = \tau_0 + \delta \tau_{ii}$$

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$D = \boldsymbol{r}_{pe} \cdot \boldsymbol{\epsilon}_I$ …

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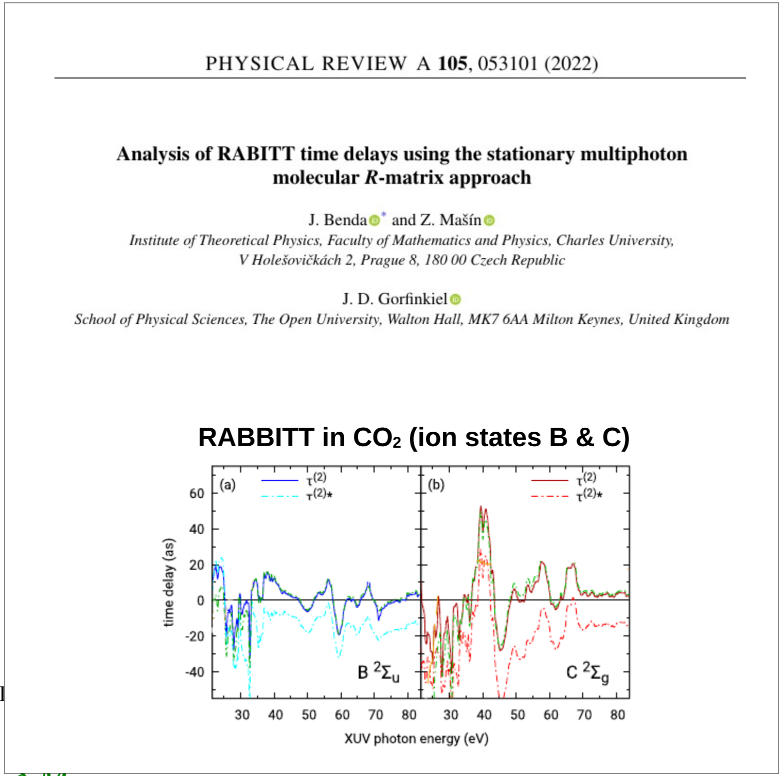
PHYSICAL REVIEW A 105, 053101 (2022)

Analysis of RABITT time delays using the stationary multiphoton molecular *R*-matrix approach

J. Benda* and Z. Mašín
Institute of Theoretical Physics, Faculty of Mathematics and Physics, Charles University, V Holešovičkách 2, Prague 8, 180 00 Czech Republic

J. D. Gorfinkiel
School of Physical Sciences, The Open University, Walton Hall, MK7 6AA Milton Keynes, United Kingdom

RABBITT in CO_2 (ion states B & C)

Asymptotic RABBITT delays

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PHYSICAL REVIEW A **105**, 053101 (2022)

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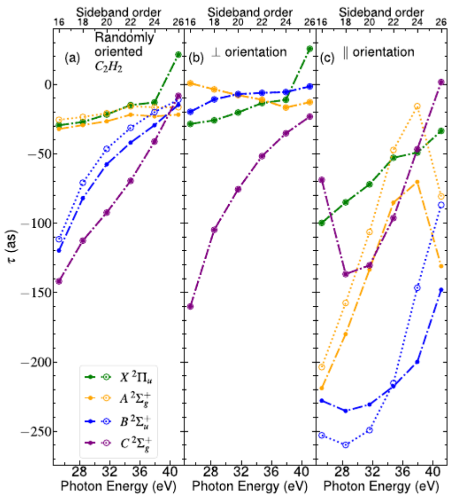
...cs, Charles University,
...ublic
...Milton Keynes, United Kingdom

PHYSICAL REVIEW A **111**, 063107 (2025)
Editors' Suggestion

Three-path interferences in the reconstruction of attosecond beatings by interference of two-photon transitions in molecules

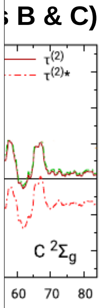
Jorge Delgado,[1,2] Celso M. González-Collado,[2] Piero Decleva,[3] Alicia Palacios,[2,4,5,*] and Fernando Martín[1,2,†]



RABBITT in C_2H_2

Dipole-coupled states: A & B

Full vs. empty circles: theory with/without A-B dipole coupling.



Asymptotic RABBITT delays

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τ_W – photoionization (Wigner) delay

τ_{cc} – continuum-continuum delay

τ_{ii} – ion-ion coupling delay

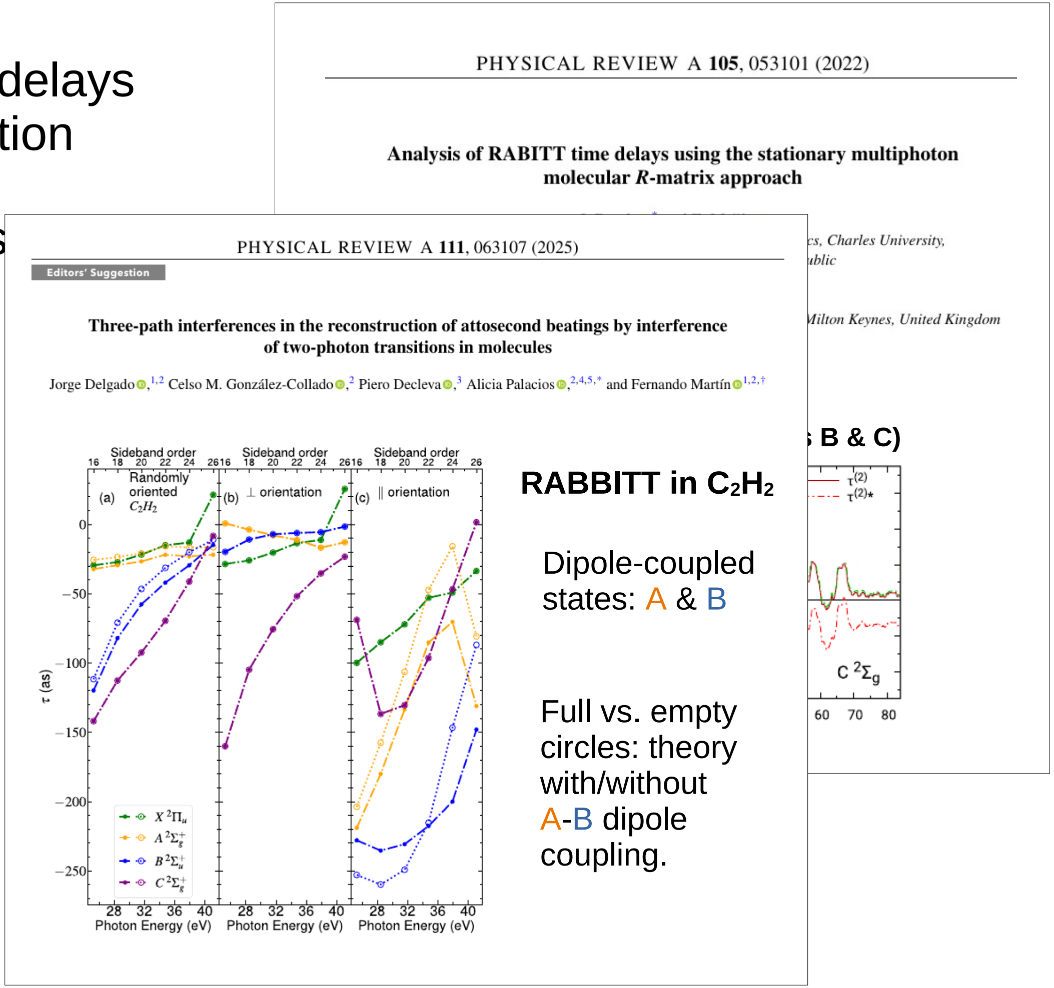
τ_{dLC} – dipole-laser coupling delay

Experimentally difficult to access

– states separated by IR quantum

– MB of one state overlaps SB of other

→ *coincidence measurement needed*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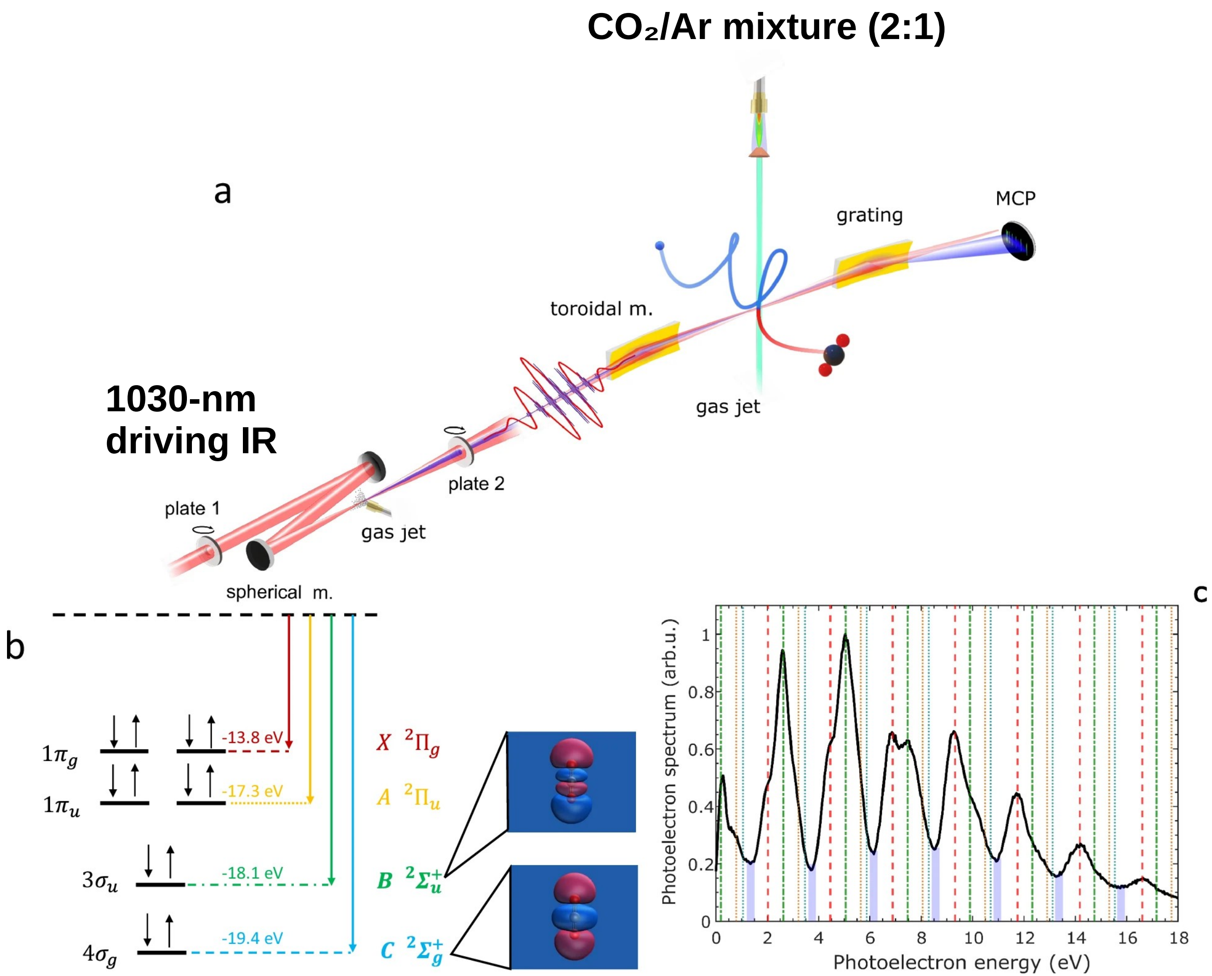
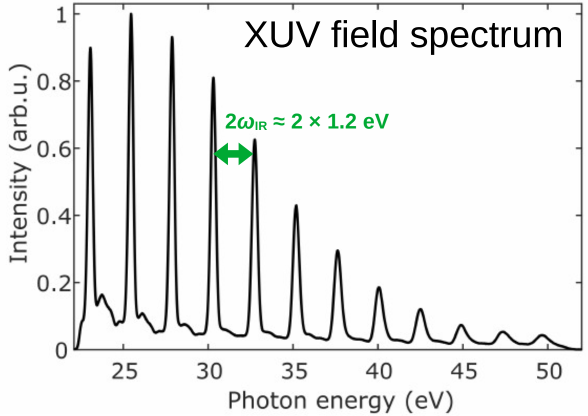
Experimental method



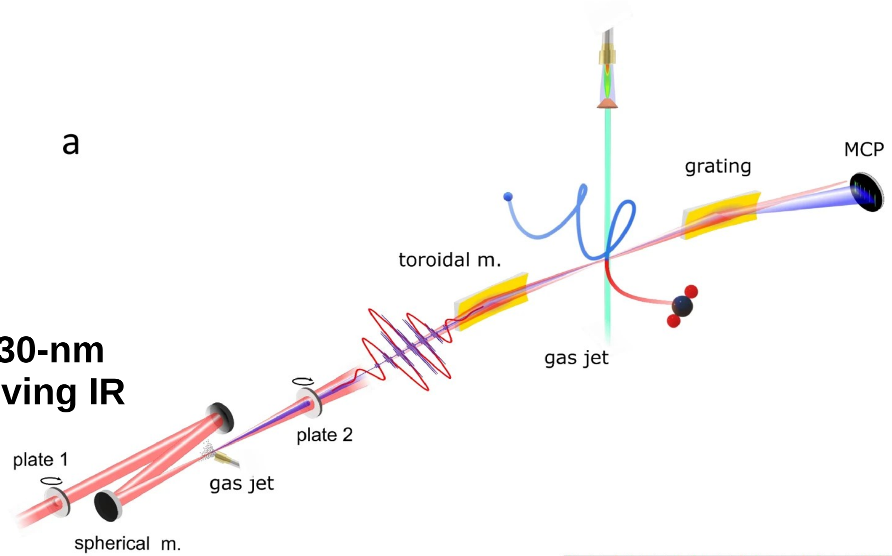


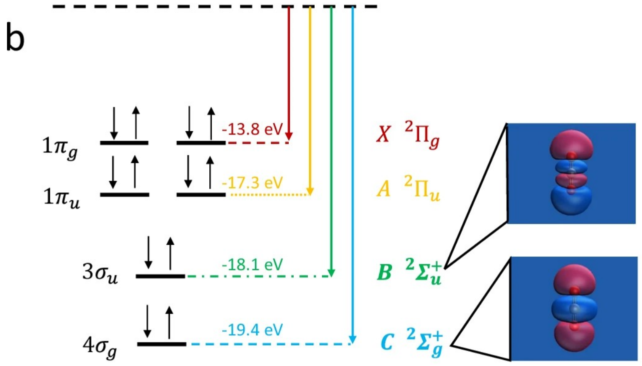
Makos et al., Nat. Commun. ***16*** *(2025) 8554*

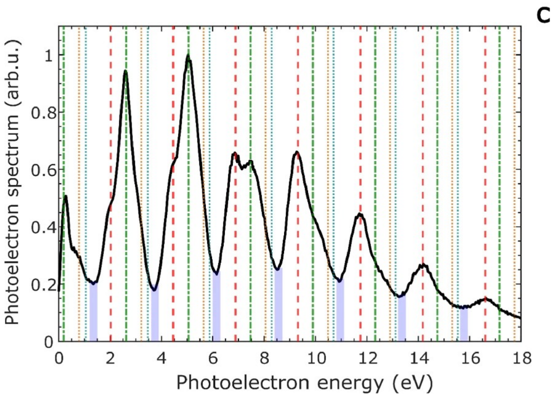
Experimental method

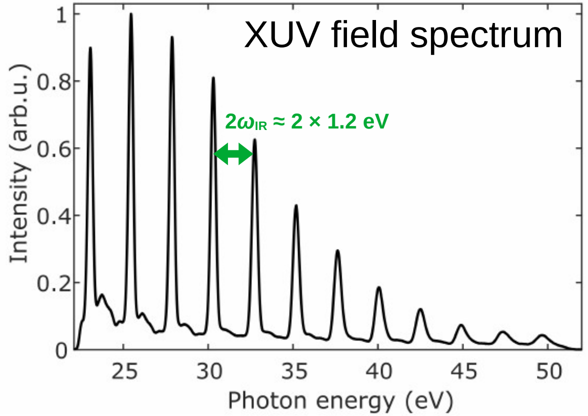
CO_2/Ar mixture (2:1)

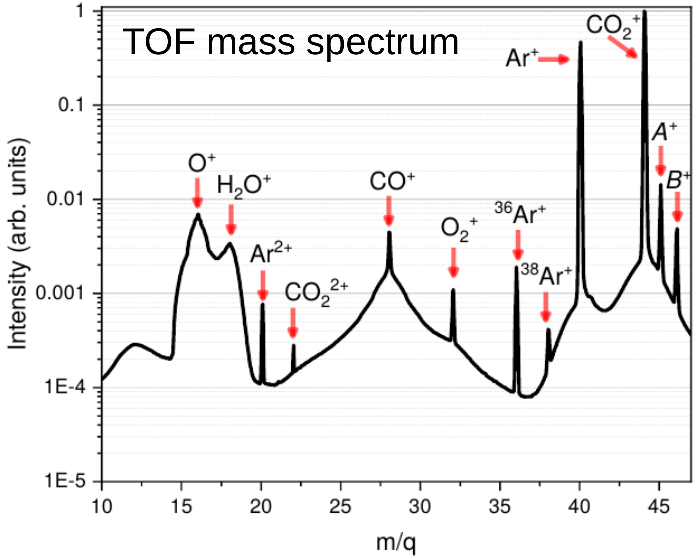
1030-nm driving IR





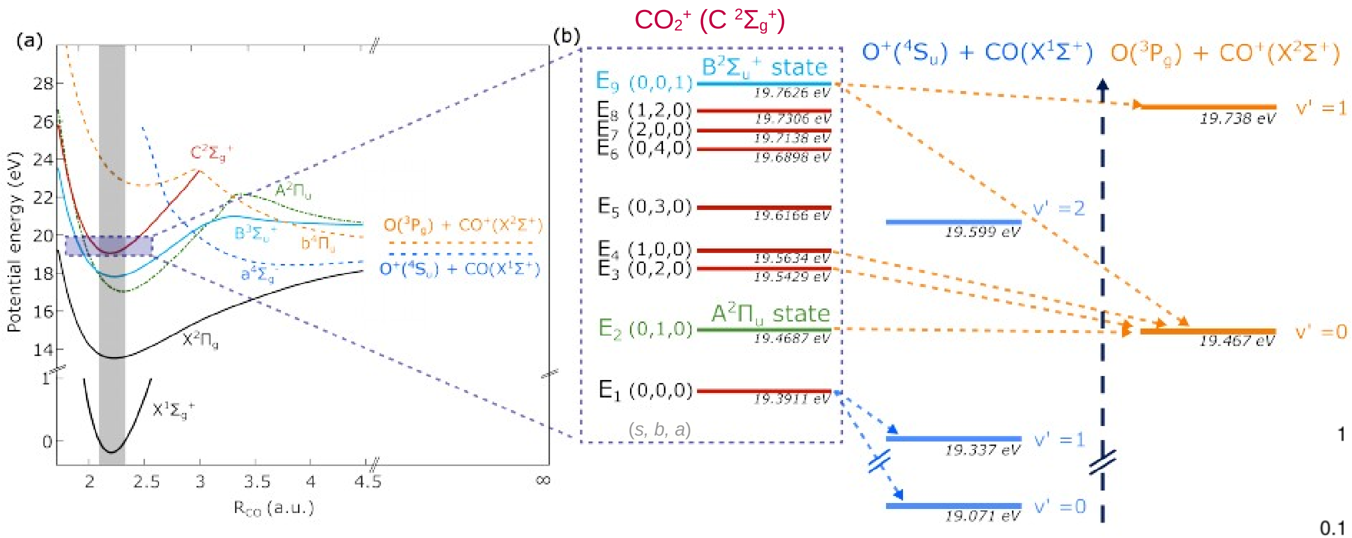


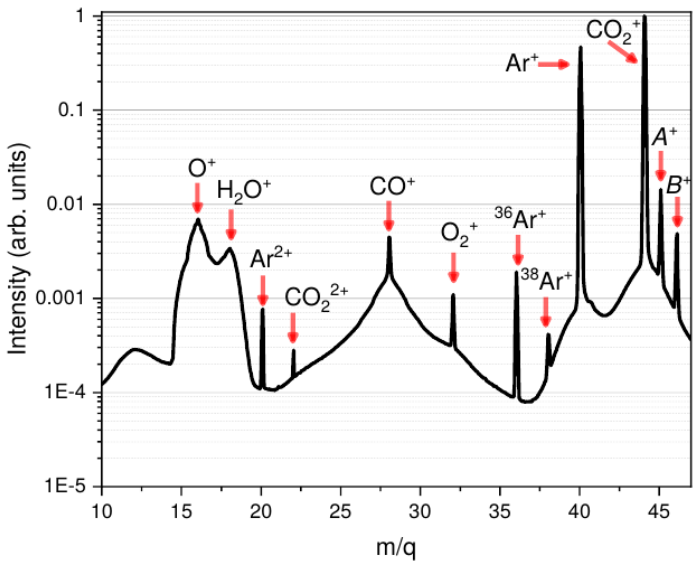




Makos et al., Nat. Commun. ***16*** *(2025) 8554*

CO_2^+ potential surfaces





Makos et al., arXiv:2604.23441

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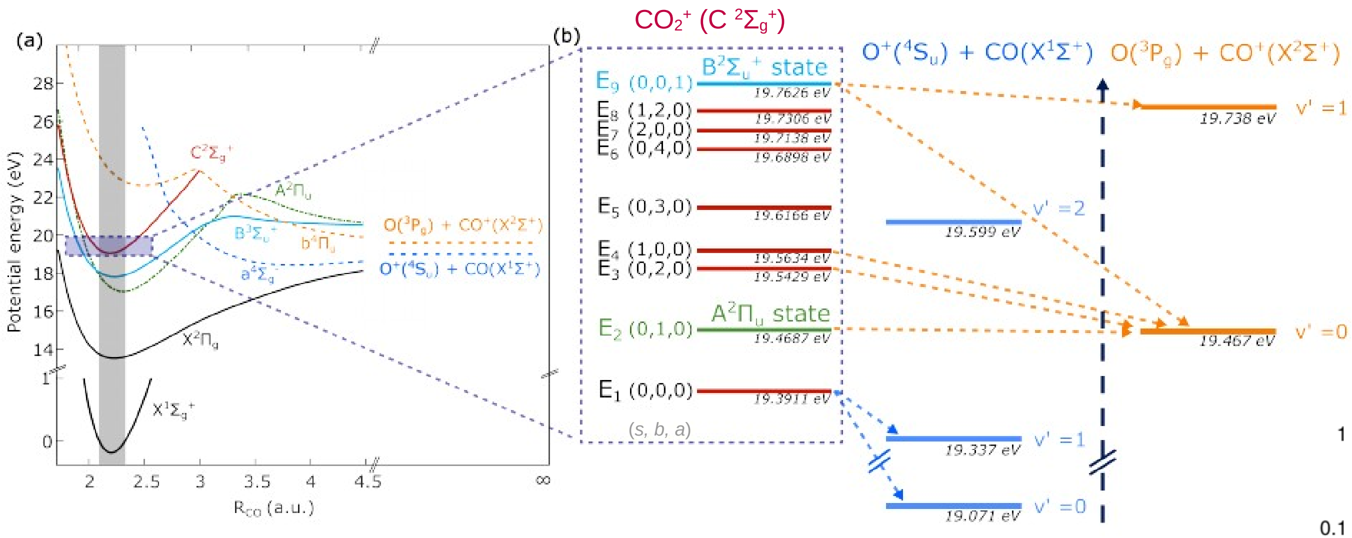
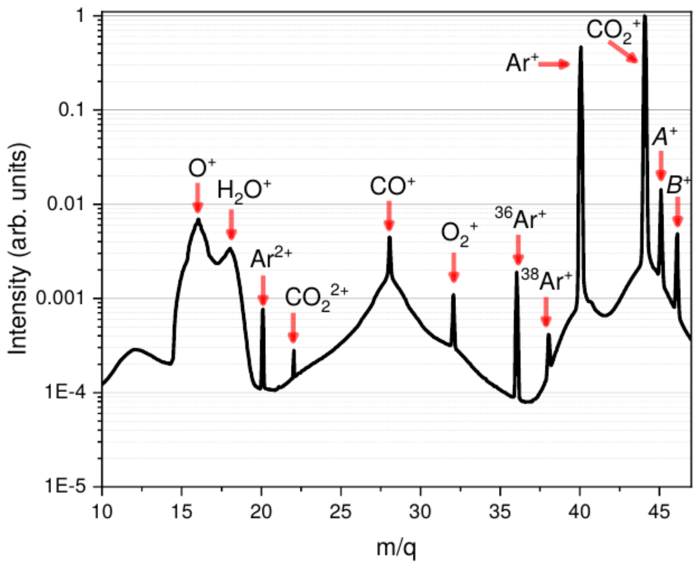


CO_2^+ B $^2\Sigma_u^+$ (ν=0) is stable



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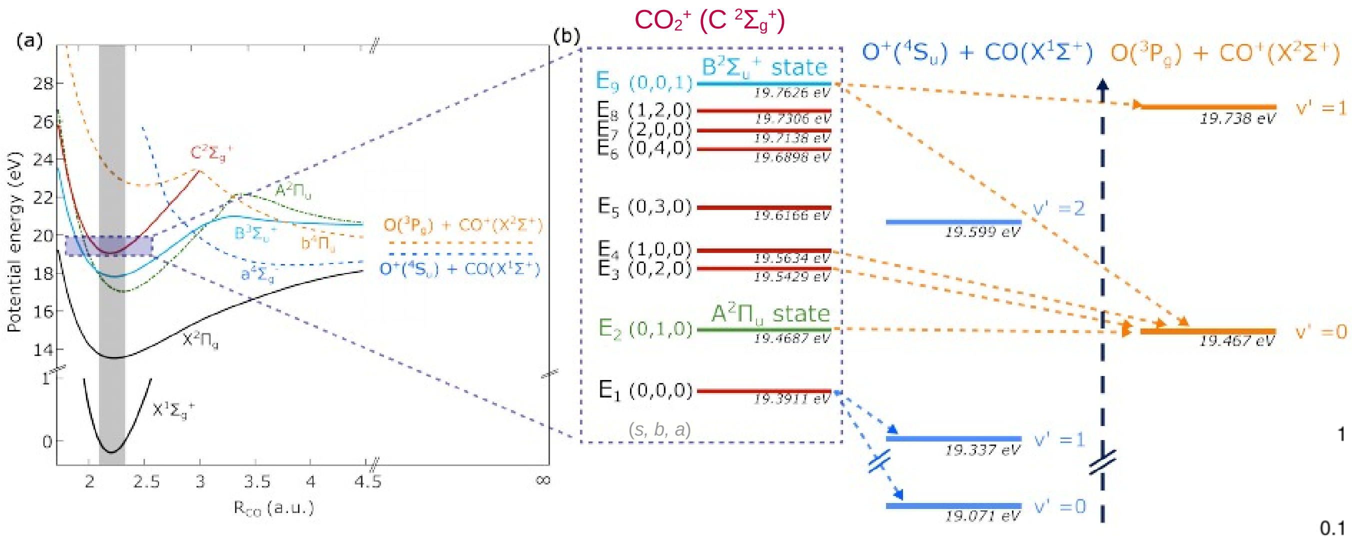

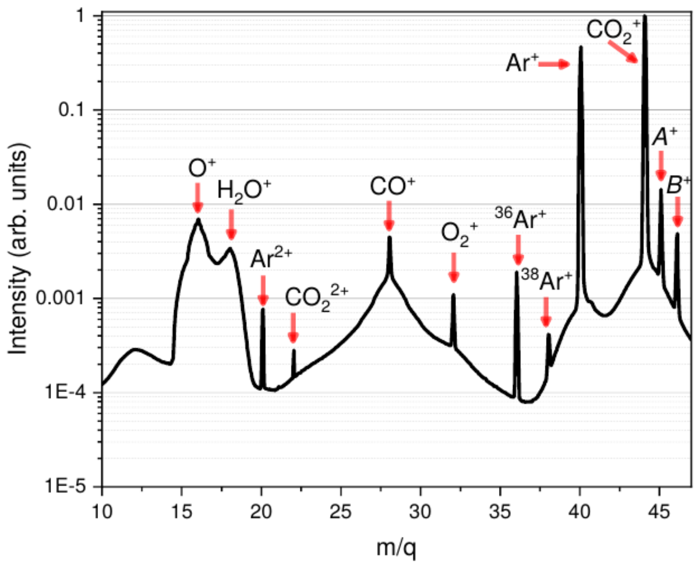

CO_2^+ $B\ ^2\Sigma_u^+$ (ν=0) is stable

CO_2^+ $C\ ^2\Sigma_g^+$ dissociates

- (ν=0): CO + O^+
- (ν>0): O + CO^+

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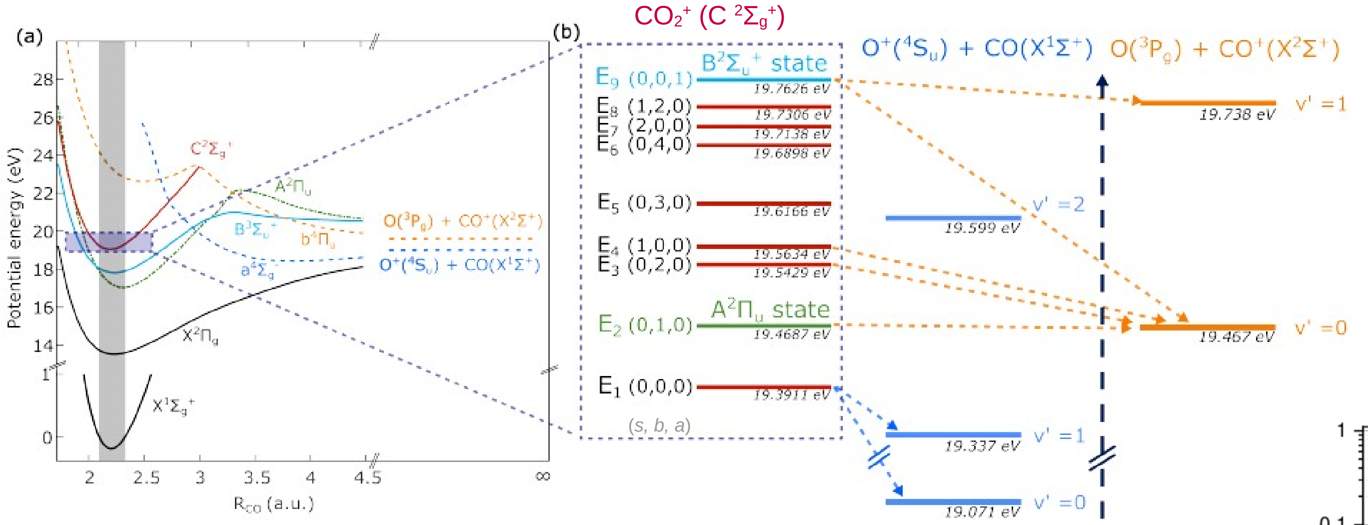

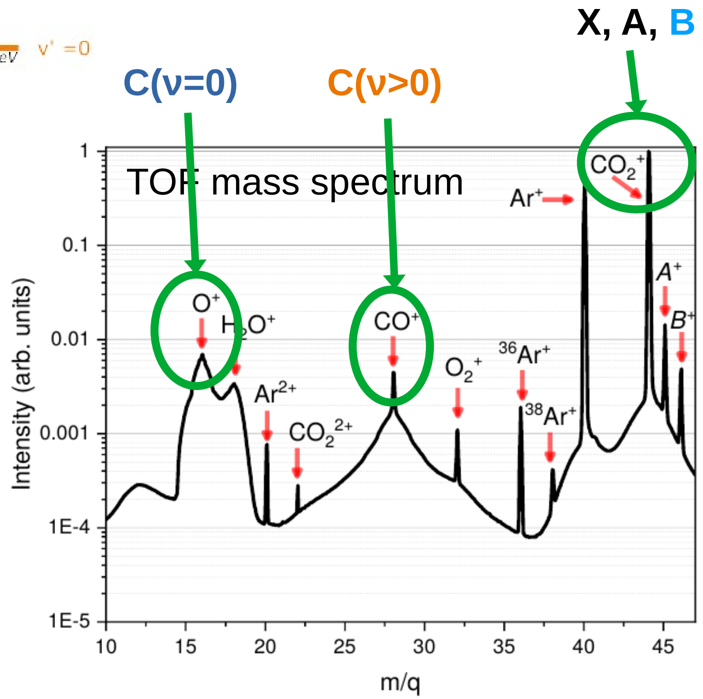

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- (ν=0): CO + O^+
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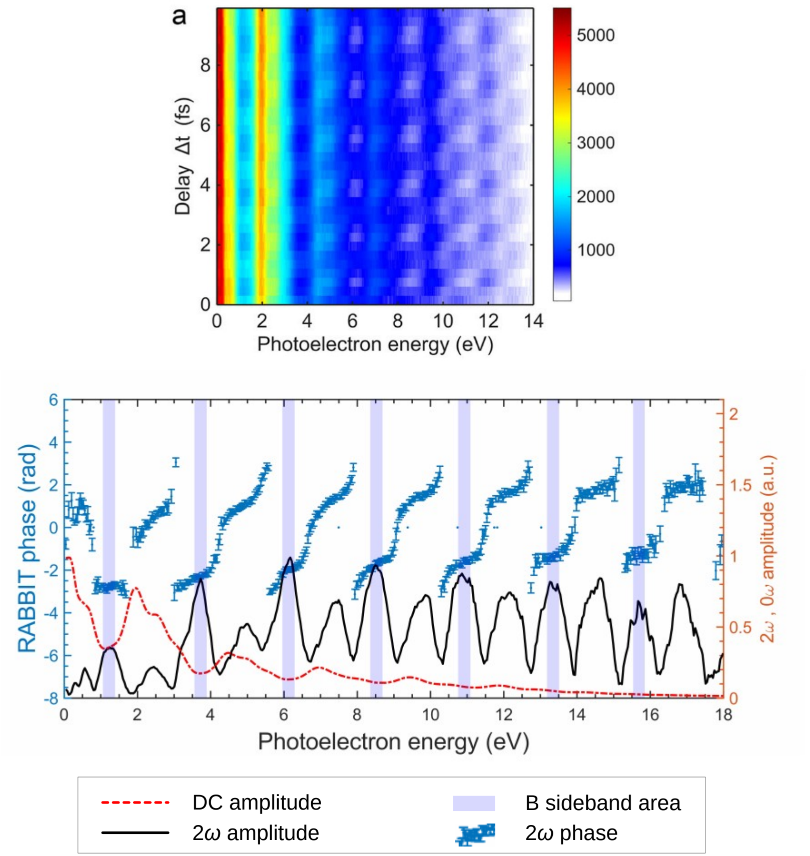
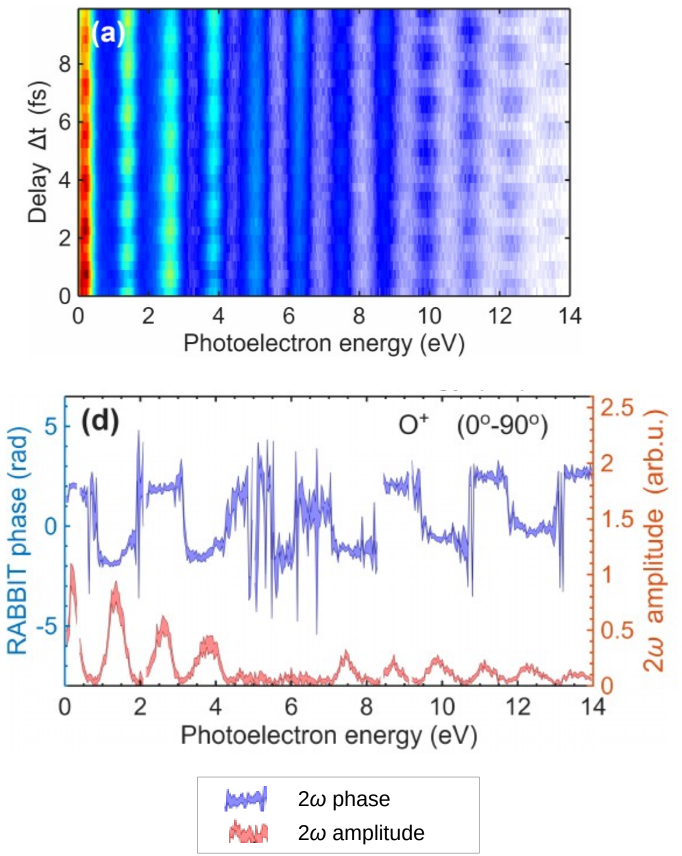
Makos et al., arXiv:2604.23441

RABBITT photoelectron spectra

In coincidence with: CO_2^+ ~ X,A,B

O^+ ~ C(ν=0)





Makos et al., Nat. Commun. **16** (2025) 8554

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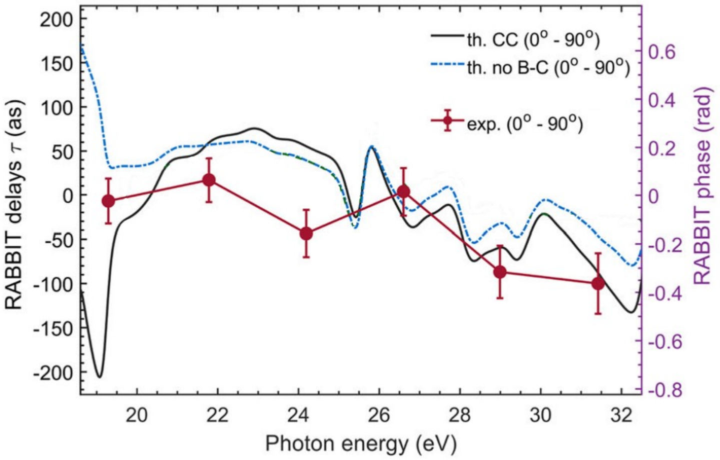
RABBITT delays

CO_2^+ **B** $^2\Sigma_u^+$, relative to $Ar^+(3p)$



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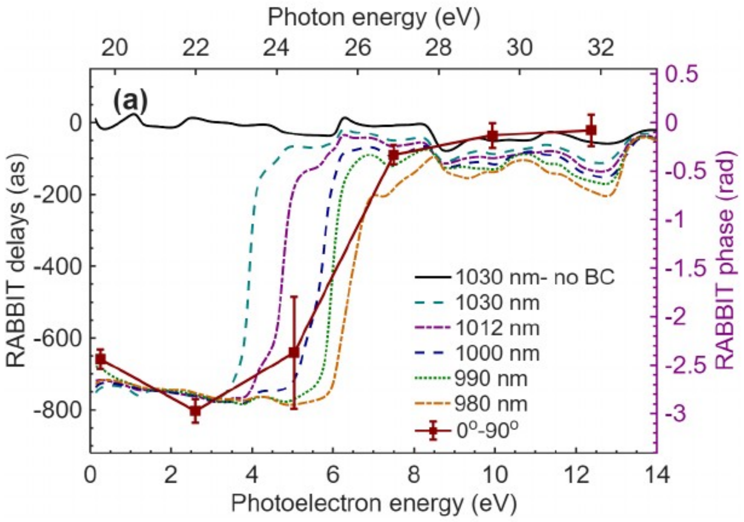

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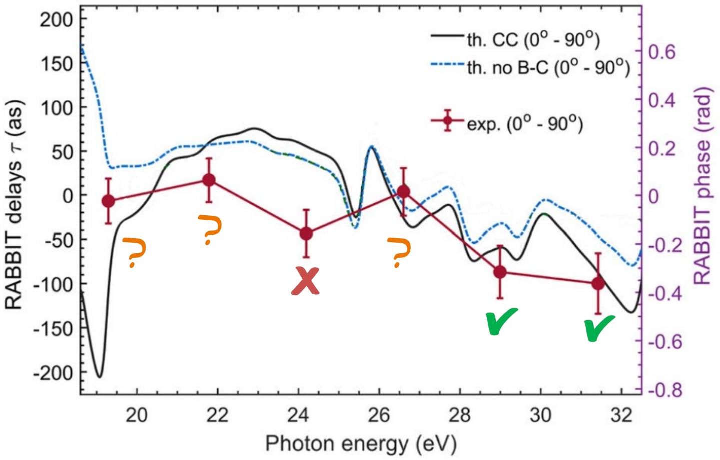

$$M = \langle \Psi_f | (\boldsymbol{r}_{\mathrm{pe}} + \boldsymbol{D}_{\mathrm{ion}}) \cdot \boldsymbol{\epsilon}_{\mathrm{IR}} | \Psi_{i+\Omega} \rangle$$

- Small effect – and nonuniform at low energies.
- Theory-experiment agreement improves towards higher energies.
- Inadequate modelling of autoionizing resonances?
- Initial-state vibrational motion?

Makos et al., Nat. Commun. ***16*** *(2025) 8554*

Pathway interference (*B* channel)

$$\tau_R = \frac{1}{2\omega_{\mathrm{IR}}} \arg M^*_{\mathrm{XUV+IR}} M_{\mathrm{XUV-IR}}$$

$$M_B = \langle \Psi_B | (\boldsymbol{r}_{\mathrm{pe}} + \boldsymbol{D}_{\mathrm{ion}}) \cdot \boldsymbol{\epsilon}_{\mathrm{IR}} | \Psi_{i+\Omega} \rangle$$

$$= M_{\mathrm{pe}} + \delta M_{\mathrm{ion}}$$

Makos et al., Nat. Commun. ***16*** *(2025) 8554*

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$$= M_{\mathrm{pe}} + \delta M_{\mathrm{ion}}$$

High-energy limit (mol. frame)

$$M_{B,\mathrm{pe}} \approx \frac{ik}{\omega^2_{\mathrm{IR}}} d_B$$

$$\delta M_{B,\mathrm{ion}} \approx \frac{1}{\Delta_{BC} - \omega_{\mathrm{IR}}} d_C \qquad \Delta_{BC} = E_C - E_B$$

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$$\rightarrow \delta\tau_{ii} \approx \frac{1}{2k} \frac{\omega_{\mathrm{IR}}}{\Delta_{BC} - \omega_{\mathrm{IR}}} \frac{\boldsymbol{D}_{BC} \cdot \boldsymbol{\epsilon}_{\mathrm{IR}}}{\boldsymbol{k} \cdot \boldsymbol{\epsilon}_{\mathrm{IR}}} \sqrt{\frac{\sigma_C}{\sigma_B}} \cos \arg d_B^* d_C$$

Makos et al., Nat. Commun. ***16*** *(2025) 8554*

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$$= M_{\mathrm{pe}} + \delta M_{\mathrm{ion}}$$

$\Rightarrow$ Coupling delay highly sensitive to relation of Δ_{BC} and ω_{IR}.

High-energy limit (mol. frame)

$$M_{B,\mathrm{pe}} \approx \frac{ik}{\omega^2_{\mathrm{IR}}} d_B$$

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$$= M_{\mathrm{pe}} + \delta M_{\mathrm{ion}}$$

⇒ Coupling delay highly sensitive to relation of Δ_{BC} and ω_{IR}.

⇒ Sign of τ_{ii} depends on sign of $\Delta_{BC} - \omega_{\mathrm{IR}}$.

High-energy limit (mol. frame)

$$M_{B,\mathrm{pe}} \approx \frac{ik}{\omega_{\mathrm{IR}}^2} d_B$$

$$\delta M_{B,\mathrm{ion}} \approx \frac{1}{\Delta_{BC} - \omega_{\mathrm{IR}}} d_C \qquad \Delta_{BC} = E_C - E_B$$

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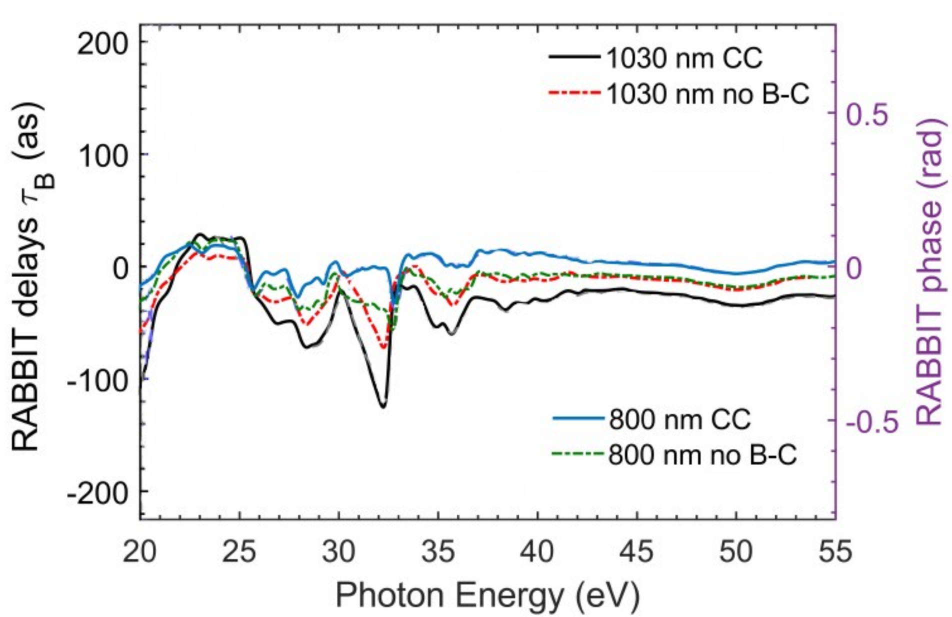


Makos et al., *Nat. Commun.* ***16*** *(2025) 8554*

Pathway interference (C channel)

$$\tau_R = \frac{1}{2\omega_{\text{IR}}} \arg M^*_{\text{XUV+IR}} M_{\text{XUV-IR}}$$

$$M_C = \langle \Psi_C | (\boldsymbol{r}_{\text{pe}} + \boldsymbol{D}_{\text{ion}}) \cdot \boldsymbol{\epsilon}_{\text{IR}} | \Psi_{i+\Omega} \rangle$$

$$= M_{\text{pe}} + \delta M_{\text{ion}}$$

⇒ Coupling delay highly sensitive to relation of Δ_{BC} and ω_{IR}.

⇒ Sign of τ_{ii} depends on sign of $\Delta_{BC} - \omega_{\text{IR}}$.

⇒ Destructive interference for comparable M_{pe} and δM_{ion}.

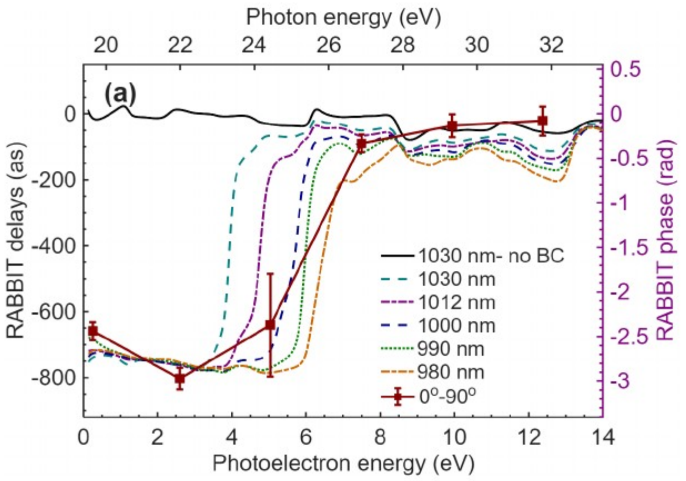
High-energy limit (mol. frame)

$$M_{C,\text{pe}} \approx \frac{ik}{\omega^2_{\text{IR}}} d_C$$

$$\delta M_{C,\text{ion}} \approx \frac{1}{\Delta_{BC} - \omega_{\text{IR}}} d_B \qquad \Delta_{BC} = E_C - E_B$$

$$\rightarrow \delta\tau_{ii} \approx \frac{1}{2k} \frac{\omega_{\text{IR}}}{\Delta_{BC} - \omega_{\text{IR}}} \ldots etc \ldots$$

CO_2^+ C $^2\Sigma_g^+$, relative to $Ar^+(3p)$



Makos et al., arXiv:2604.23441

Pathway interference (C channel)

$$\tau_R = \frac{1}{2\omega_{\mathrm{IR}}} \arg M^*_{\mathrm{XUV+IR}} M_{\mathrm{XUV-IR}}$$

$$M_C = \langle \Psi_C | (\boldsymbol{r}_{\mathrm{pe}} + \boldsymbol{D}_{\mathrm{ion}}) \cdot \boldsymbol{\epsilon}_{\mathrm{IR}} | \Psi_{i+\Omega} \rangle$$

$$= M_{\mathrm{pe}} + \delta M_{\mathrm{ion}}$$

- ⇒ Coupling delay highly sensitive to relation of Δ_{BC} and ω_{IR}.
- ⇒ Sign of τ_{ii} depends on sign of $\Delta_{BC} - \omega_{\mathrm{IR}}$.
- ⇒ Destructive interference for comparable M_{pe} and δM_{ion}.

High-energy limit (mol. frame)

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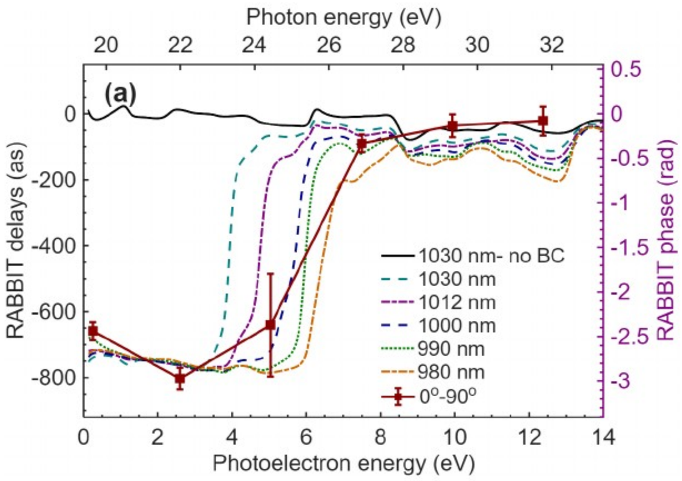
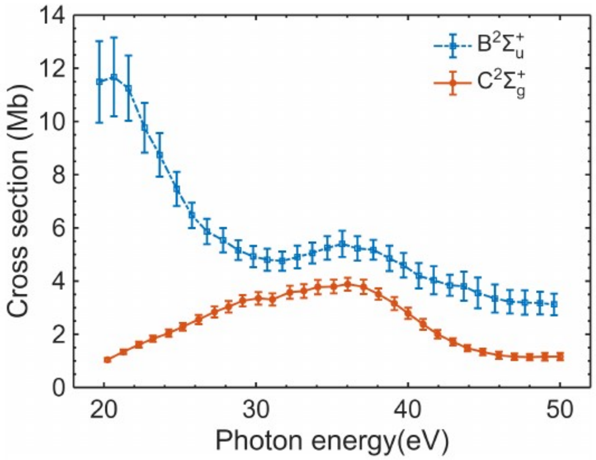


Cross sections B vs C



Makos et al., arXiv:2604.23441

Three-path RABBITT

Transition in residual ion affects delay of photoelectrons…?

Makos et al., Nat. Commun. ***16*** *(2025) 8554*

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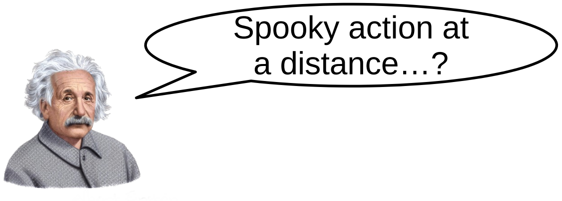

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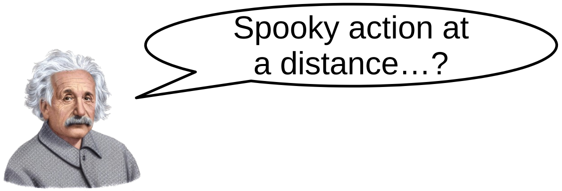
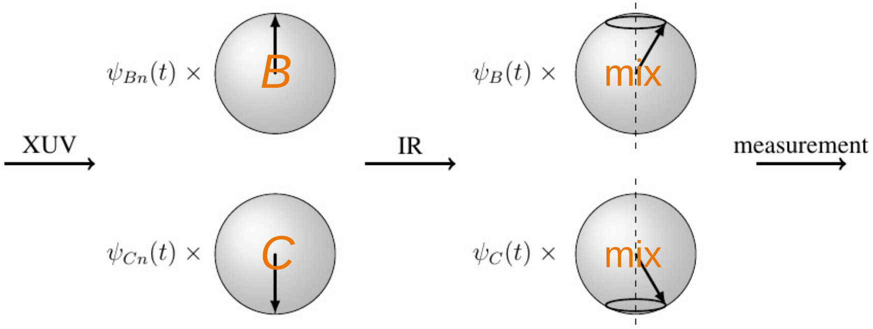


Actually: *entanglement*



Projection on *B* or *C* (after IR) mixes photoelectrons associated with the original *B* and *C* channels.

Makos et al., Nat. Commun. ***16*** *(2025) 8554*

Entanglement

Degree of entanglement affects delay of photoelectrons

Makos et al., Nat. Commun. ***16*** *(2025) 8554*

Degree of entanglement affects delay of photoelectrons

State after XUV ionization: $\Psi_n \sim \psi_B^{\text{pe}} \Phi_B^{\text{ion}} + \psi_C^{\text{pe}} \Phi_C^{\text{ion}}$ $\qquad \rho = |\Psi_n\rangle\langle\Psi_n|$

⇩

Reduced DM of the ion subsystem: $\rho_{\text{ion}} = Tr_{\text{pe}}\, \rho \sim \frac{1}{\sigma_B + \sigma_C} \begin{pmatrix} \sigma_B & 0 \\ 0 & \sigma_C \end{pmatrix}$

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Purity of the ion subsystem: $\gamma_{\text{ion}} = Tr\, \rho_{\text{ion}}^2 \sim \frac{\sigma_B^2 + \sigma_C^2}{(\sigma_B + \sigma_C)^2}$

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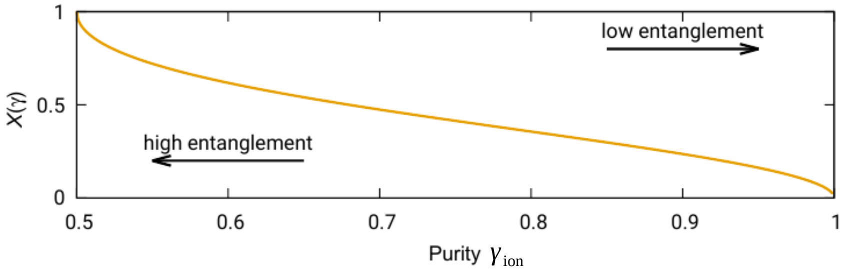
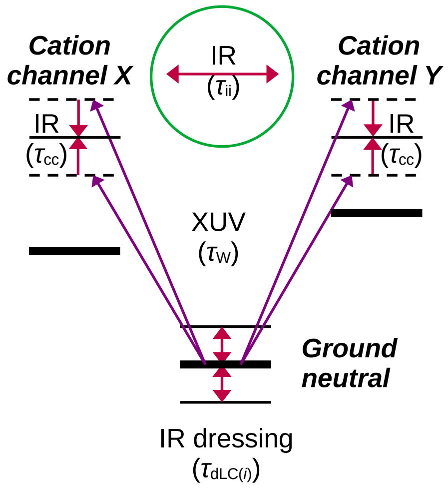


⇒ Coupling delay high for *highly entangled* intermediate states.

Makos et al., *Nat. Commun.* ***16*** *(2025) 8554*

Conclusion

- IR-driven transitions in residual ion affect RABBITT delays.



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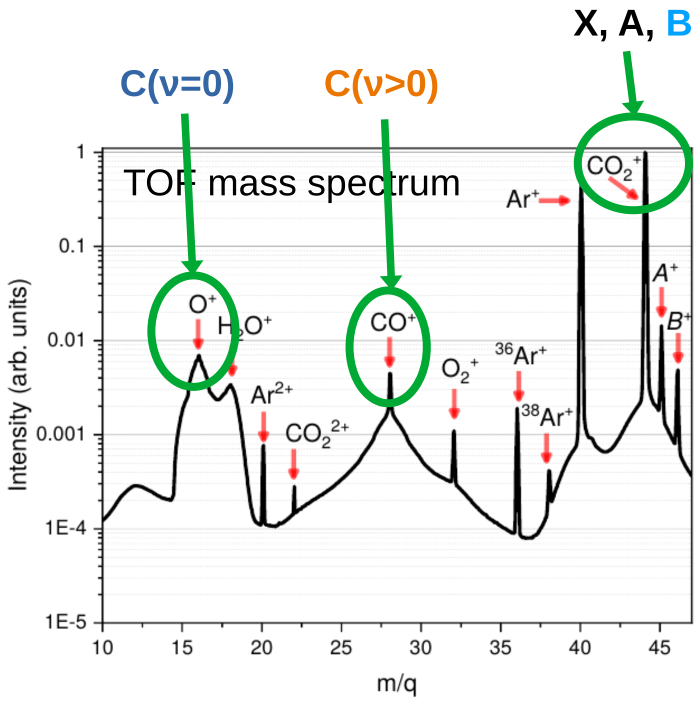

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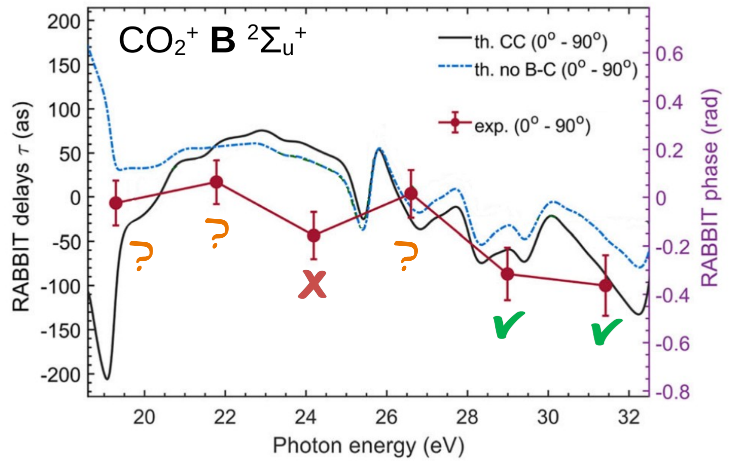

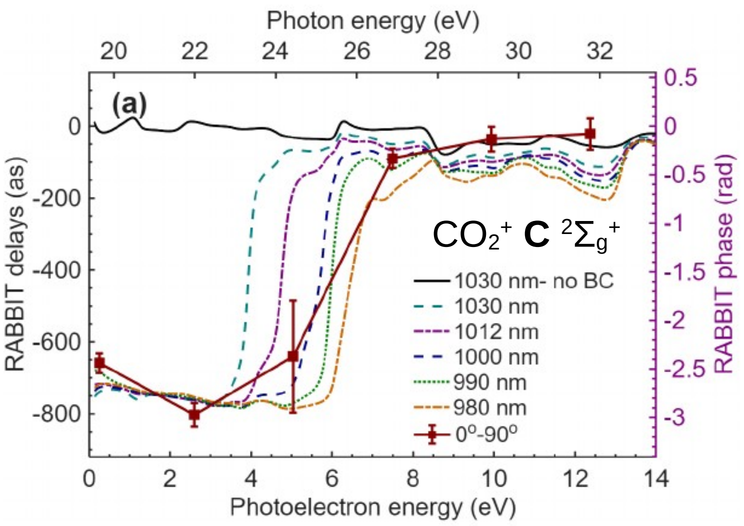

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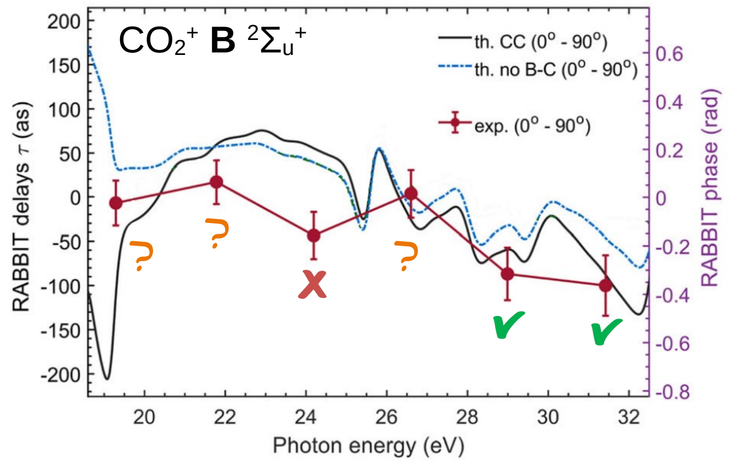

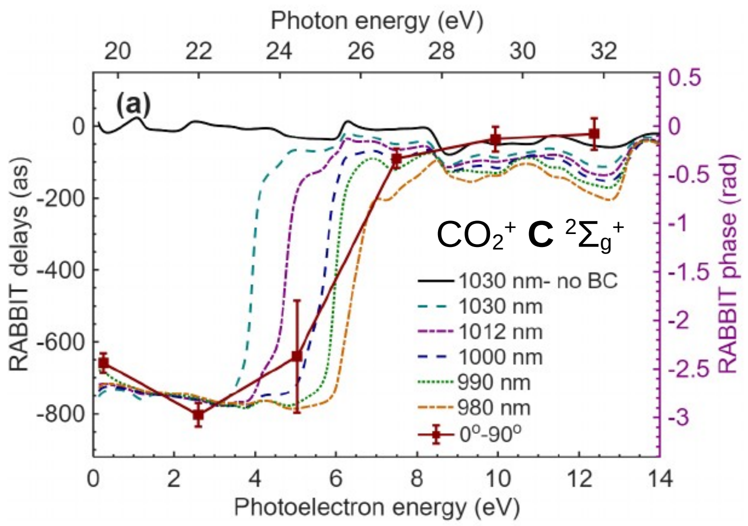

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Thank you for attention!