Multi-photon above-threshold ionization of atoms and molecules using the time-independent R-matrix approach

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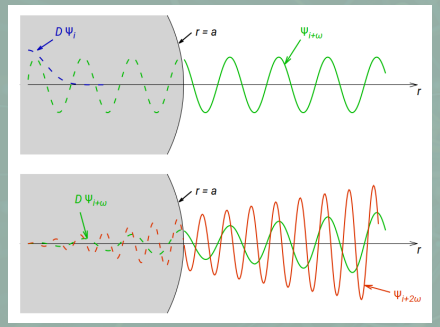
Method: R-matrix division of space

Inner region ($r < a$)
- many-electron wave function expanded in box eigenstates
- boundary condition to match outer solutions

Outer region ($r > a$)
- 1-electron channel expansion
- central Coulomb potential of ion dominates over multipoles
- analytic procedures (Coulomb, Whittaker, Green's functions)



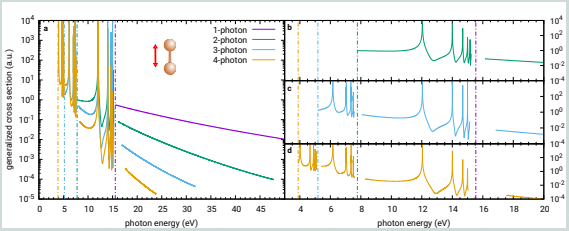
Solution of intermediate states

$$(E_i + \omega - H)\,\Psi^{(+)}_{i+\omega} = D_1\,\Psi_i$$
$$(E_i + 2\,\omega - H)\,\Psi^{(+)}_{i+2\,\omega} = D_2\,\Psi^{(+)}_{i+\omega}$$

use intermediate state in the next order

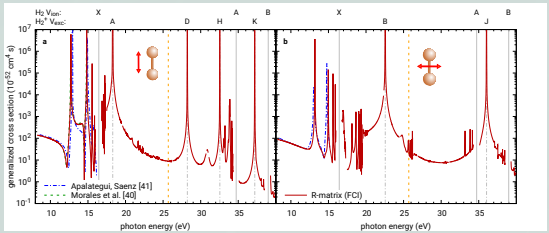
- embed the boundary condition "inner region generates outgoing wave H^+ only"
- evaluate outer free-free integrals using asymptotic form of Coulomb functions

Multi-photon ionization of H_2 in static exchange (single channel) model, including ATI for parallel orientation of molecular axis and field polarization. Multi-photon thresholds are marked by chain lines. Resonances correspond to intermediate excitations of the neutral molecule.

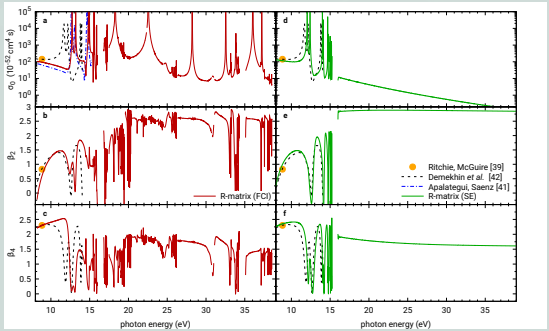


Results: Two-photon cross sections

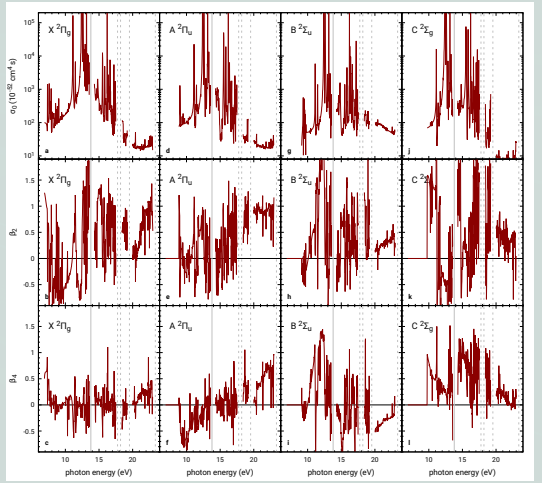
Two-photon ionization of H_2 by field polarized parallel (left panel) and perpendicular (right panel) to the molecular axis compared to other calculations. Grey lines mark one- (solid) and two-photon (broken) thresholds. The yellow line marks the double ionization threshold. Full CI model with aug-cc-pVTZ basis set was used.



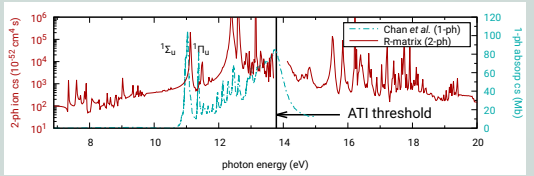
Parameters of molecular-orientation-averaged photoelectron angular momentum distribution (total isotropic cross section σ_0 and asymmetry parameters β_2 and β_4 for the first ionic state) after **two-photon ionization of H_2** compared to other calculations. Results obtained with the full CI model (left) as well as in the static exchange approximation (right) are presented.



Parameters of molecular-orientation-averaged photoelectron angular momentum distribution (partial isotropic cross section σ_0 and asymmetry parameters β_2 and β_4) after **two-photon ionization of CO_2** into one of the four lowest states **X, A, B, C**. Molecular orbitals constructed from the basis set cc-pVTZ and optimized in CASSCF were used, together with a CAS model of CO_2 that involved 300 ionic states.



Below-threshold **two-photon cross section of CO_2** compared to one-photon absorption cross section measurement for corresponding resonance features.



References

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Mašín *et al.*, J. Phys. B: At. Mol. Opt. Phys. **51** (2018) 134006 ... source of the molecular model of CO_2 used in this work
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