Ab initio study of the ellipticity of molecular high harmonic generation driven by linearly polarized laser fields

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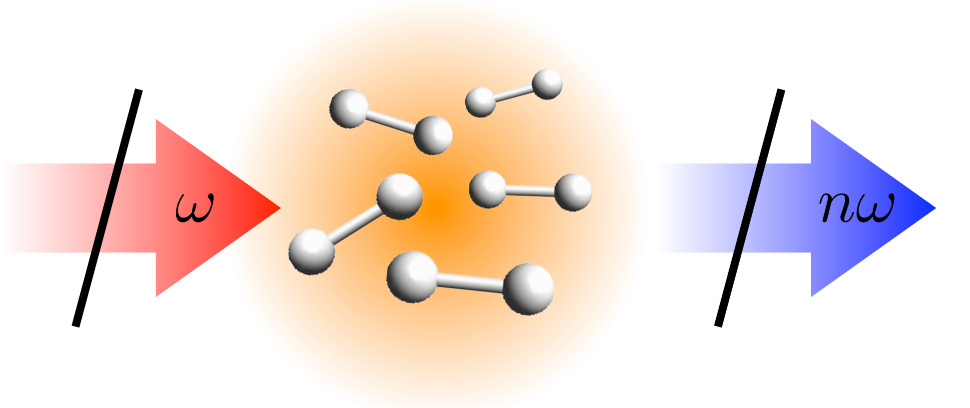
Abstract

A recent experiment has demonstrated that high-order harmonic generation (HHG) from aligned linear molecules can be elliptically polarized even if driven by linearly polarized laser fields [Zhou *et al.* (2009)]. We perform fully *ab initio* calculations of HHG from the ground and excited electronic states of H_2^+ with arbitrary orientation and detailed analyses for the polarization and phases of harmonic emissions to reveal theoretical origins of the ellipticity of molecular HHG. Our results predict that even for the one-electron system all harmonic emissions are elliptically polarized unless molecular alignment is parallel or perpendicular to the polarization of the driving laser field. The ellipticity of harmonic emissions is closely related to the symmetry of the molecular orbital and affected by two-center interference effects in HHG. For H_2^+, the ellipticity becomes large for the ground state that is approximated by a symmetric combination of the atomic orbitals, whereas it becomes small for the first excited state approximated by an antisymmetric combination. This observation can be generalized for the ellipticity of HHG from linear molecules.

Elliptical HHG

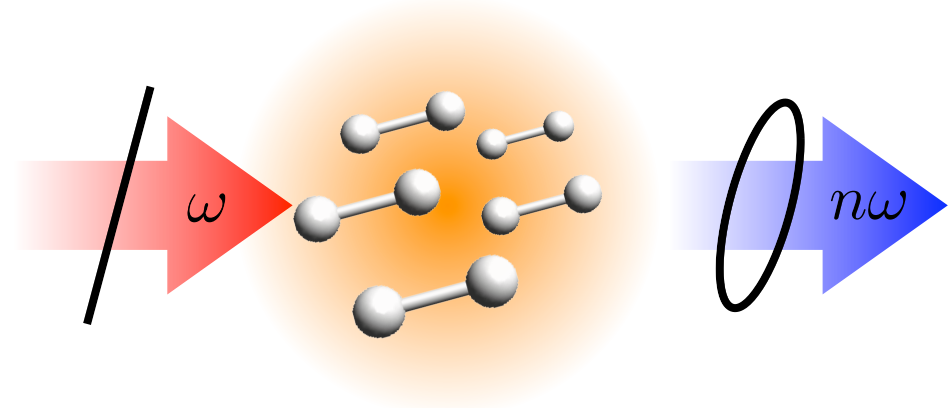
For atoms or unaligned molecules



linearly polarized driving laser field

linearly polarized HHG

For aligned linear molecules



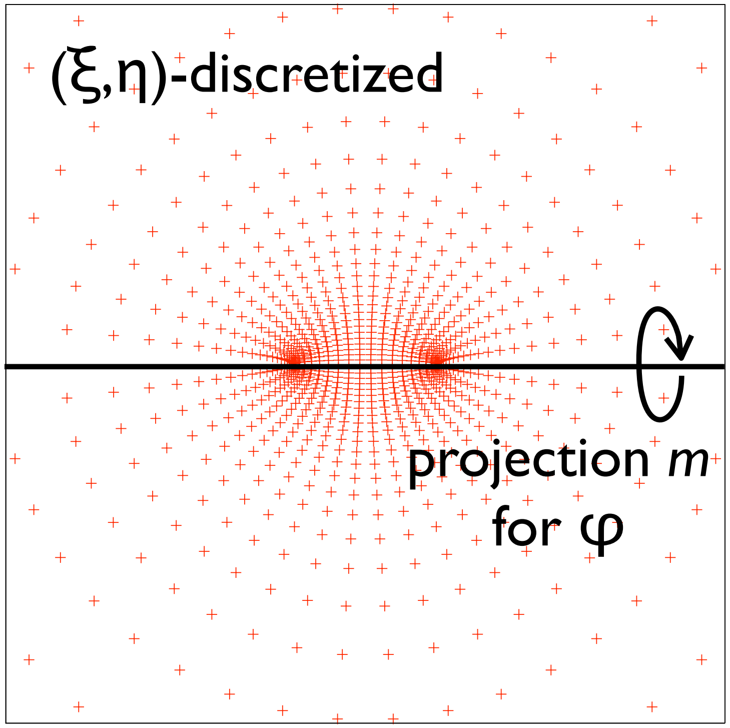
linearly polarized driving laser field

elliptically polarized HHG

- Levesque *et al.* (2007) and Lee *et al.* (2008): no ellipticity but tilted linearity in experiment
- Zhou *et al.* (2009): high ellipticity for N_2 but no ellipticity for CO_2 in experiment
- Smirnova *et al.* (2009): high ellipticity for CO_2 in theory (multiple elec. continuum dynamics)
- Etches *et al.* (2010): unexpectedly small ellipticity in theory (modified SFA)
- Ramakrishna *et al.* (2010): possibility of ellipticity in theory (modified SFA)

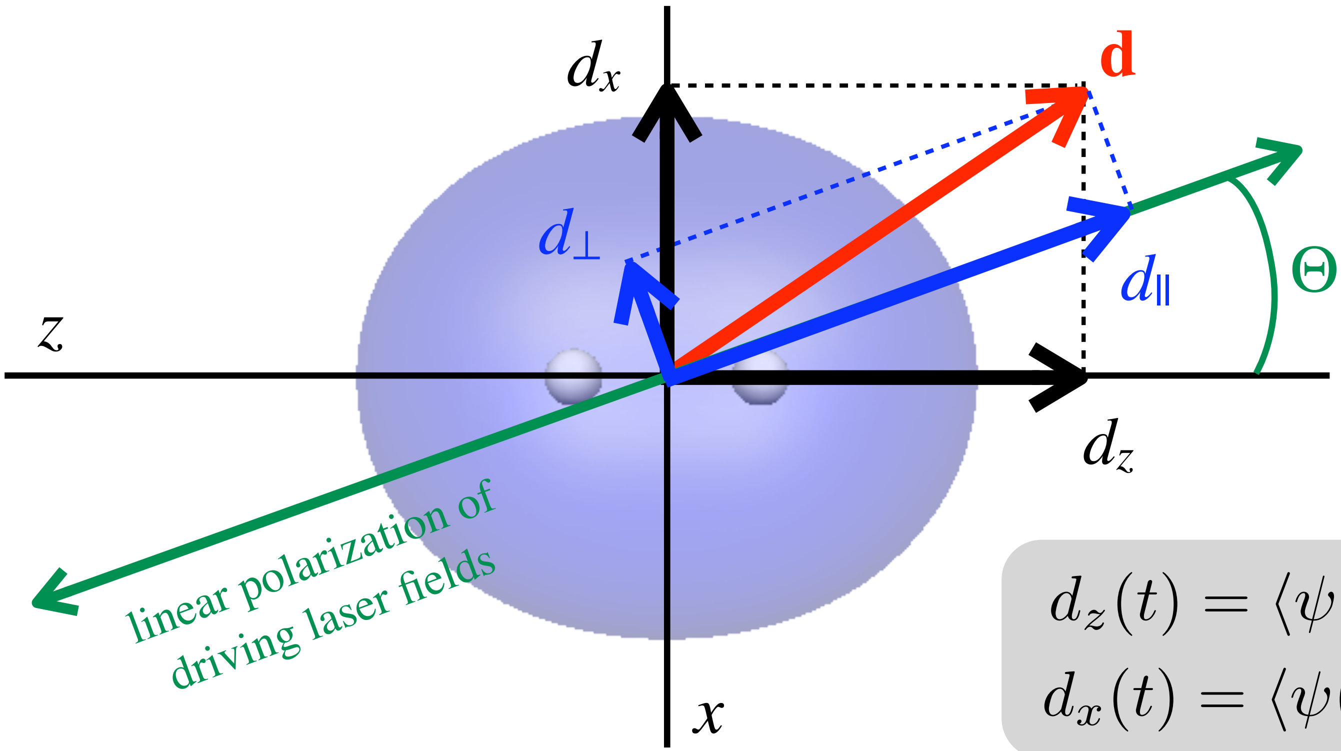
Numerical method

- *ab initio* and high-precision calculation of 3D TDSE
- A two-center problem of H_2^+ is accurately and efficiently solved by the time-dependent generalized pseudospectral (TDGPS) method in the prolate spheroidal coordinates [Telnov & Chu (2007)].



non-uniform and optimal grids for H_2^+ in the prolate spheroidal coordinates

Two components of dipole moment



$$d_z(t) = \langle \psi(t) | z | \psi(t) \rangle$$
$$d_x(t) = \langle \psi(t) | x | \psi(t) \rangle$$

parallel component:
perpendicular component:

$$d_\parallel(t) = d_z(t) \cos\Theta + d_x(t) \sin\Theta$$
$$d_\perp(t) = d_x(t) \cos\Theta - d_z(t) \sin\Theta$$

(w.r.t. the driving laser field polarization)

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Ellipticity

Spectral dipole:

$$\tilde{d}_{\|/\perp}(\omega) = \int_{-\infty}^{\infty} d_{\|/\perp}(t) e^{i\omega t} dt$$

$$= \left| \tilde{d}_{\|/\perp}(\omega) \right| e^{i\phi_{\|/\perp}(\omega)}$$

Amplitude:

$$A_{\|/\perp}(\omega) = \sqrt{\frac{4\omega^4}{6\pi c^3}} \left| \tilde{d}_{\|/\perp}(\omega) \right|$$

Phase:

$\phi_{\|/\perp}(\omega)$

Ellipticity:

$$\varepsilon = \sqrt{\frac{1 + r^2 - \sqrt{1 + 2r^2 \cos 2\delta + r^4}}{1 + r^2 + \sqrt{1 + 2r^2 \cos 2\delta + r^4}}}$$

where $r = A_\perp / A_\|$ and $\delta = \phi_\perp - \phi_\|$

- $\varepsilon = 0$: linear
- $0 < \varepsilon < 1$: elliptic
- $\varepsilon = 1$: circular

For high ellipticity, both conditions must be satisfied: i) $r \approx 1$ and ii) $\delta \approx \pm\pi/2$

Orientation dependence of HHG

- For the parallel component, the orientation dependence is well explained by *the two-center interference model*. The harmonic amplitude has an extremum in the orientation dependence according to the symmetry of the wave function.

  i) symmetric combination of atomic orbitals → min. $A_{||}$

  ii) antisymmetric combination of atomic orbitals → max. $A_{||}$

- For the perpendicular component, the orientation dependence is qualitatively described by *the first-order perturbation theory*.

$$d_{\perp}(t) = \frac{1}{2}(\alpha_x - \alpha_z)E(t)\sin 2\Theta \quad \rightarrow \text{always maximum } A_{\perp}$$

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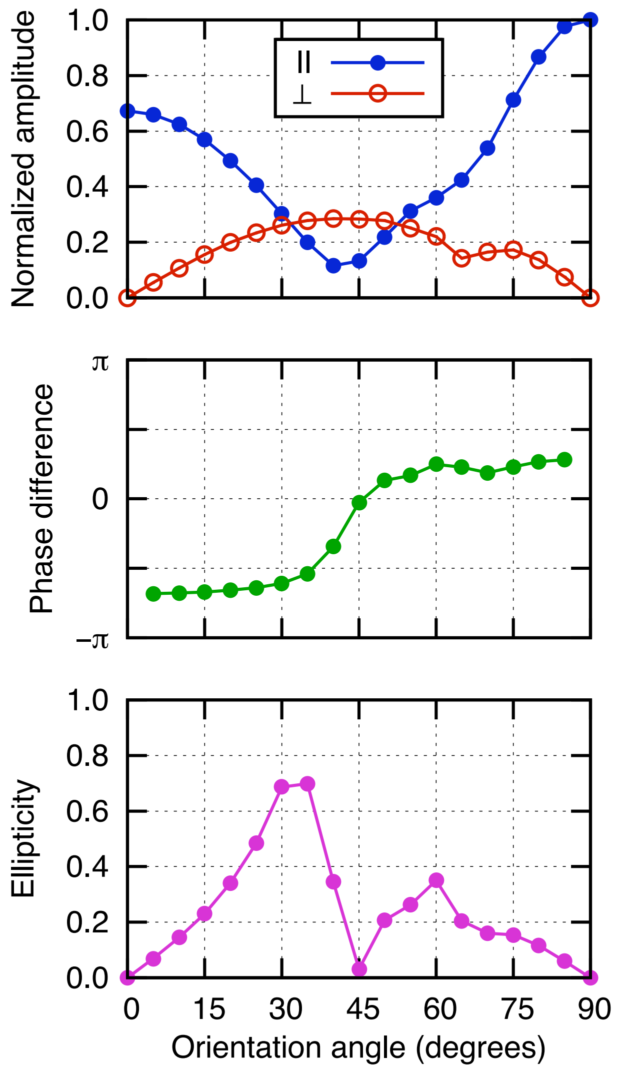
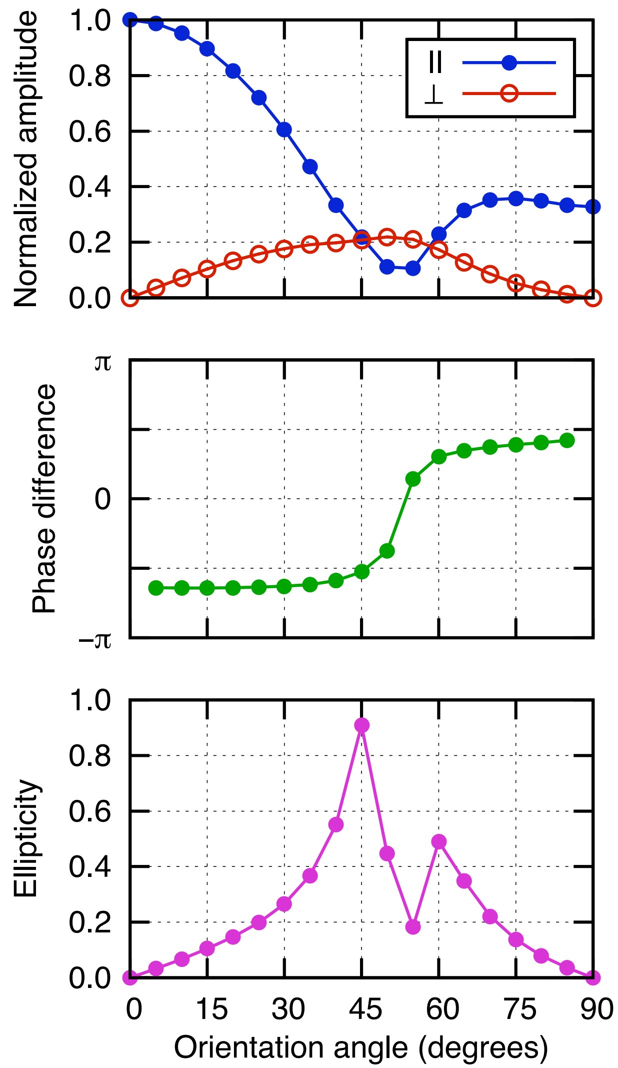
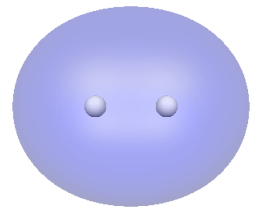
Symmetric 1σ_g state





H_2^+ 1σ_g



symmetric combination of atomic orbitals

↓

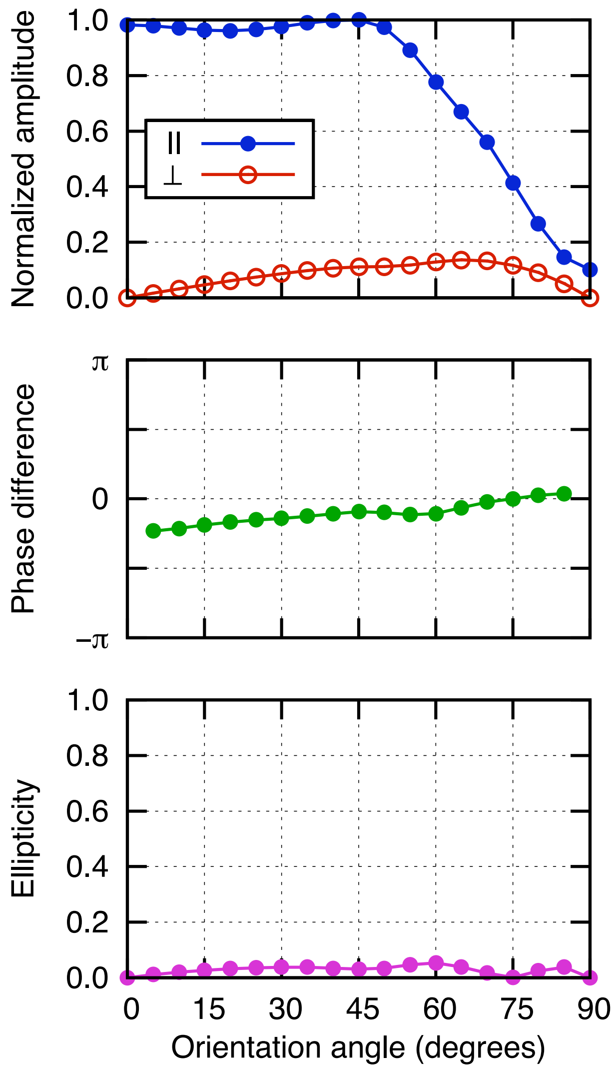
minimum $A_{||}$

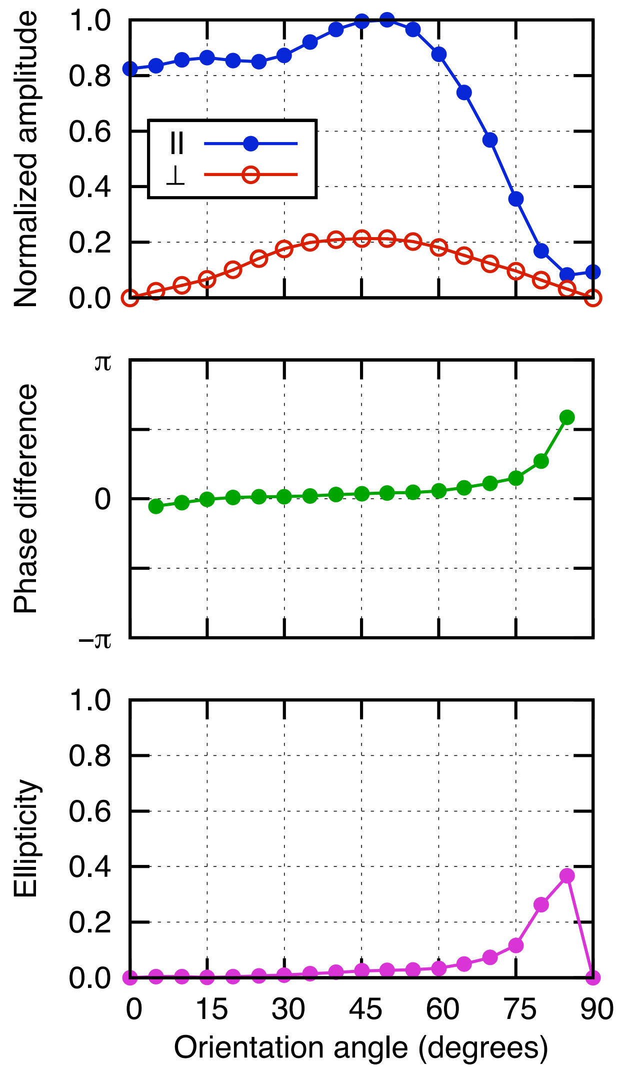
maximum $A_{\perp}$

↓

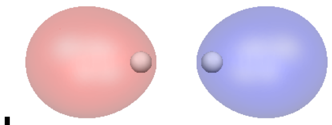
high ellipticity

Antisymmetric 1σ_u state





H_2^+ 1σ_u



antisymmetric combination of atomic orbitals

↓

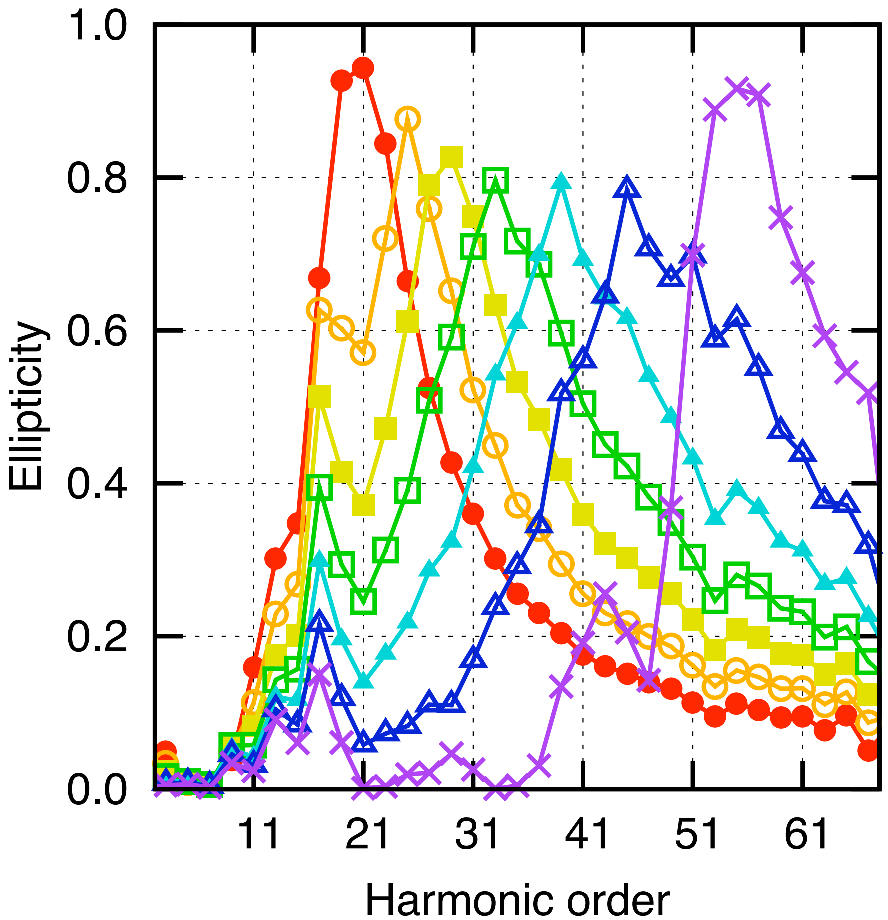
maximum $A_{||}$

maximum $A_{\perp}$

↓

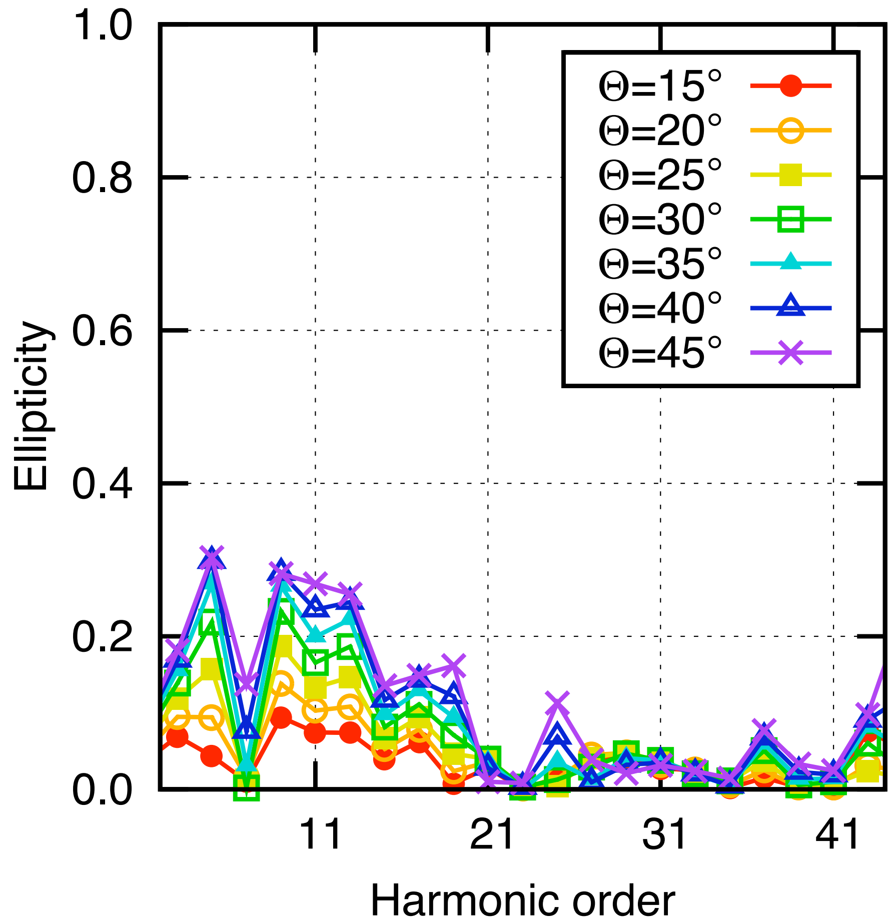
low ellipticity

Ellipticity of H_2^+



symmetric $1\sigma_g$

at 800 nm & 3×10^{14} W/cm^2



antisymmetric $1\sigma_u$

at 800 nm & 2×10^{14} W/cm^2

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Conclusion

- Theoretical origins of the elliptical HHG from aligned linear molecules in the linearly polarized strong laser field have been revealed by means of high-precision *ab initio* calculations.
- All harmonic radiations can be elliptically polarized unless the molecular alignment is parallel or perpendicular to the driving laser polarization.
- Elliptical HHG is observed even without any multielectron effects.
- We propose an instructive prediction of the ellipticity of molecular HHG related to the molecular orbital symmetry: high for symmetric combination of atomic orbitals but low for antisymmetric combination of atomic orbitals.

References

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