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Non-dipole Effects in Multi-Photon Ionization of the Hydrogen Atom using Intense Laser Pulses

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Introduction and Motivation

The temporal and spatial variation of the laser has a great bearing on above threshold ionization (ATI) spectrum and the ionization rate [1]. For laser fields with lower intensities and/or longer wavelengths, the spatial distribution may be ignored and the use of dipole approximation is justified. But for high intensity and/or very short wavelength laser sources currently available, the excursion radius r of an atomic, ionic or molecular system in the radiation field may be comparable to the wavelength λ. This would make the spatial dependence of the laser field non-negligible and consequently, the dipole (Dip) approximation may not be valid. In this case, the simulation of the electron dynamical processes of the system needs to include the spatial distribution of the laser field.

In this study, the non-dipole (ND) effects arising from the spatial variation are investigated upto the quadrupole term of the multipole expansion. We were able to reproduce literature data [2] which were also evaluated by spectral expansion using B splines. We confirm the existence of significant non-dipole effects at all intensities corresponding with the peak electric field strengths considered in that work.

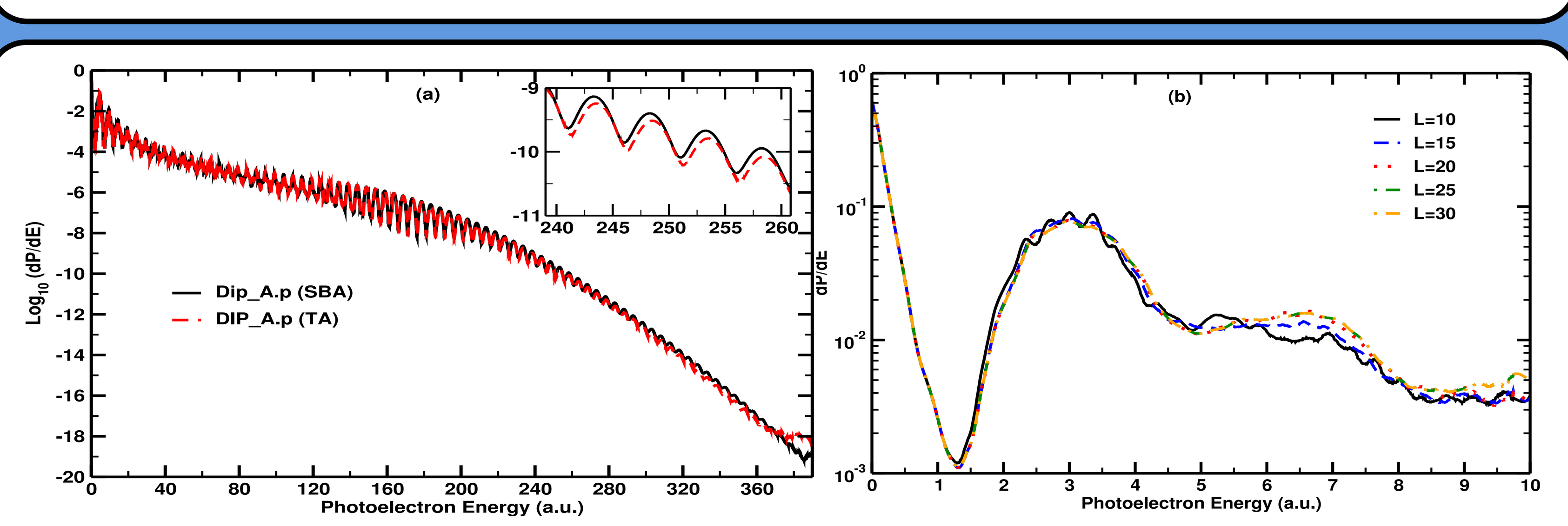

Figure 1. (a) Comparison of the Taylor approximated(TA) and the sherical Bessel approximated (SBA) dipole PE spectrum at a peak electric field strength of 50 au for 9.11 nm 10 cycle pulse. Inset: Minor spatial modulation of the PE spectrum at higher energy regime. (b) Convergence of dipole plus electric quadrupole PE spectrum with angular momentum at a peak electric field strength of 45 au for a 13 nm 15 cycle laser pulse.

Theory and Method

- Non-relativistic time-dependent Schrödinger equation (TDSE) of an atom interacting with a classical electromagnetic field

$$i\frac{\partial\Psi(\mathbf{r},t)}{\partial t} = [\hat{\mathrm{H}}_0 + \hat{\mathrm{H}}_{int}(\mathbf{r},t)]\,\Psi(\mathbf{r},t)$$

- Field-free Hamiltonian $\hat{\mathrm{H}}_0$ and the interaction potential $\hat{\mathrm{H}}_{int}(\mathbf{r},t)$ in radiation gauge:

$$\hat{\mathrm{H}}_0 = \frac{1}{2}\mathbf{p}^2 + \mathrm{V}(r)$$
$$\hat{\mathrm{H}}_{int}(\mathbf{r},t) = -q\mathbf{A}\cdot\mathbf{p} + \frac{1}{2}q^2\mathbf{A}^2$$

- Vector potential

$$\begin{aligned}\mathbf{A}(\mathbf{r},t) &= A_0\, f(t)\, \sin(\omega t - \mathbf{k}\cdot\mathbf{r} + \delta)\,\hat{z}\\ &= A_1(t)\cos(\mathbf{k}\cdot\mathbf{r}) - A_2(t)\sin(\mathbf{k}\cdot\mathbf{r})\,\hat{z}\\ &= A(t)\cos(\mathbf{k}\cdot\mathbf{r}) + \frac{E(t)}{\omega}\sin(\mathbf{k}\cdot\mathbf{r})\,\hat{z}\end{aligned}$$

- Time-dependent potential functions

$$\begin{aligned}A_1(t) &= A_0\, f(t)\, \sin(\omega t + \delta)\\ A_2(t) &\approx A_0\, f(t)\, \cos(\omega t + \delta)\\ &= A_0\,[\frac{\dot{f}(t)}{\omega}\,\sin(\omega t + \delta) + f(t)\,\cos(\omega t + \delta)]\end{aligned}$$

- Spatial dependence in Taylor (TA) multipole expansion

$$\begin{aligned}e^{i\mathbf{k}\cdot\mathbf{r}} &= \cos(\mathbf{k}\cdot\mathbf{r}) + i\sin(\mathbf{k}\cdot\mathbf{r})\\ &= \sum_{l=0}^{\infty}\frac{(i\mathbf{k}\cdot\mathbf{r})^l}{l!}\end{aligned}$$

- Spatial dependence in spherical Bessel functions (SBA) multipole expansion $j_l(kr)$ [3]

$$e^{i\mathbf{k}\cdot\mathbf{r}} = 4\pi\sum_{l=0}^{\infty}\sum_{m_l=-l}^{+l} i^l j_l(kr) Y_l^{m*}(\hat{\mathbf{k}}) Y_l^m(\hat{\mathbf{r}})$$

$$\cos(\mathbf{k}\cdot\mathbf{r}) = 4\pi\sum_{l,m=0,2,\cdots}^{\infty} i^l\, j_l(kr)\, Y_l^{m*}(\hat{\mathbf{k}})\, Y_l^m(\hat{\mathbf{r}})$$

$$\sin(\mathbf{k}\cdot\mathbf{r}) = 4\pi\sum_{l,m=1,3,\cdots}^{\infty} i^{(l-1)}\, j_l(kr)\, Y_l^{m*}(\hat{\mathbf{k}})\, Y_l^m(\hat{\mathbf{r}})$$

- Ansatz spectrally expanded in field-free states

$$\Psi(\mathbf{r},t) = \sum_{\alpha=\{n,l,m\}} C_\alpha(t)\,\phi_\alpha(\mathbf{r})$$
$$\phi_\alpha(\mathbf{r}) = R_{n_\alpha,l_\alpha}(r)\,Y_{n_\alpha,l_\alpha}(\hat{\mathbf{r}})$$
$$R_{n_\alpha,l_\alpha}(r) = \sum_i c_i\frac{B_i(r)}{r}$$

- TDSE expanded as a set of coupled first-order ordinary differential equations

$$i\dot{C}_\alpha(t) = C_\alpha(t)\,E_\alpha + \sum_{\alpha'} C_{\alpha'}(t)\,V_{\alpha\alpha'}(t)$$
$$V_{\alpha\alpha'}(t) = \langle\phi_\alpha|\,\hat{\mathrm{H}}_{int}(\mathbf{r},t)\,|\phi_{\alpha'}\rangle$$

- Ionization yield is defined as

$$Y_{\text{ion}} = \sum_{\alpha=\text{all continuum states}} |\,C_\alpha(t=\tau)\,|^2$$

where τ is the total duration of the laser pulse.

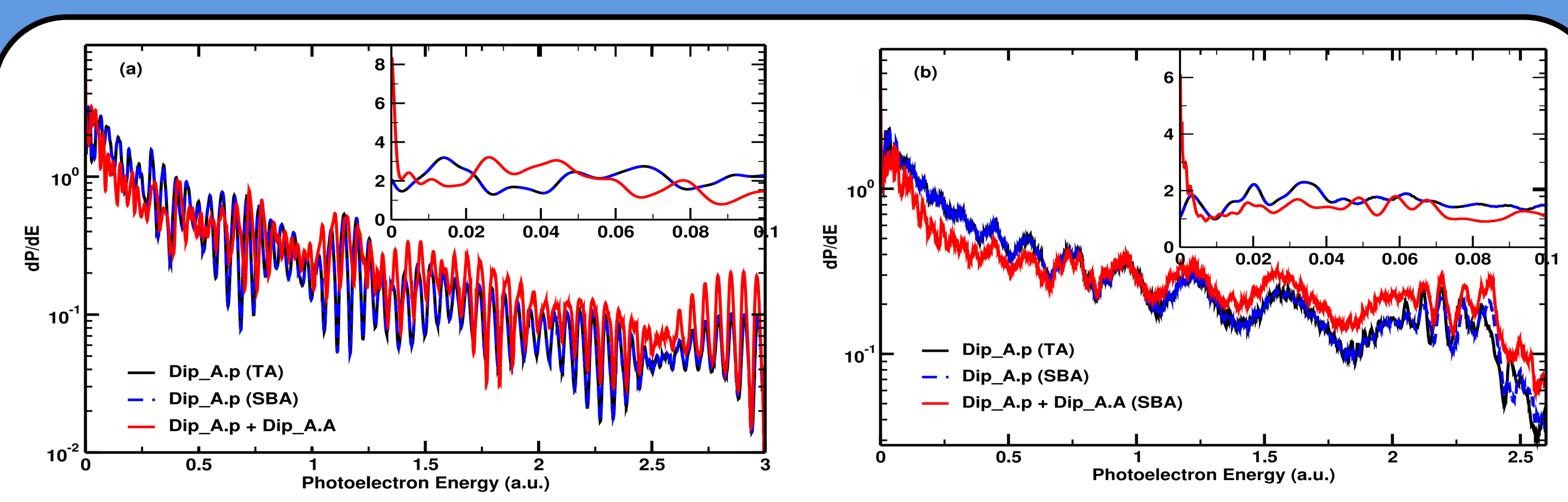

Figure 2. PE spectrum at an intensity of (a) 1.0×10^{15} Wcm^{-2} and (b) 1.0×10^{16} Wcm^{-2} for 800 nm 10 cycle pulse. Used: $r_{max} = 2000$ au, B splines= 4000, $L_{max} = 50$. Ponderomotive shift U_p significantly modifies the PE spectrum. Inset: Shows the very low energy structures "(VLES)" enhanced by this ponderomotive shift.

Acknowledgement

DAAD Deutscher Akademischer Austausch Dienst / Servicio Alemán de Intercambio Académico
Bundesministerium für Bildung und Forschung
XLIC XUV/X-ray light and fast ions for ultrafast chemistry
COST European Cooperation in Science and Technology
Technical University of Mombasa

Results

- System Parameters : Hydrogen atom, Initial state 1s.
- Laser Parameters: Wavelength $= 9.11$, 13, and 800nm. Peak electric field strengths $= 25, 35, 45$, and 50 au for short wavelengths, ~ 1 au for 800nm. Pulse duration $= 5, 10, 15$ optical cycles, linear polarization in the $\hat{z}$ direction.
- Numerical Parameters: Box radius $\mathrm{r_{max}} = 200$ au, B splines= 600, Gauge= Radiation, Angular momenta $\mathbf{L_{max}}, \mathbf{M_{max}} = 30$, Pulse envelope $= \mathrm{Cos}^2$, Approx. order $=$ quadrupole.
- Definitions: Taylor Approximation (TA), Spherical Bessel functions Approximation (SBA). Unless specified SBA is used throughout in our calculations.

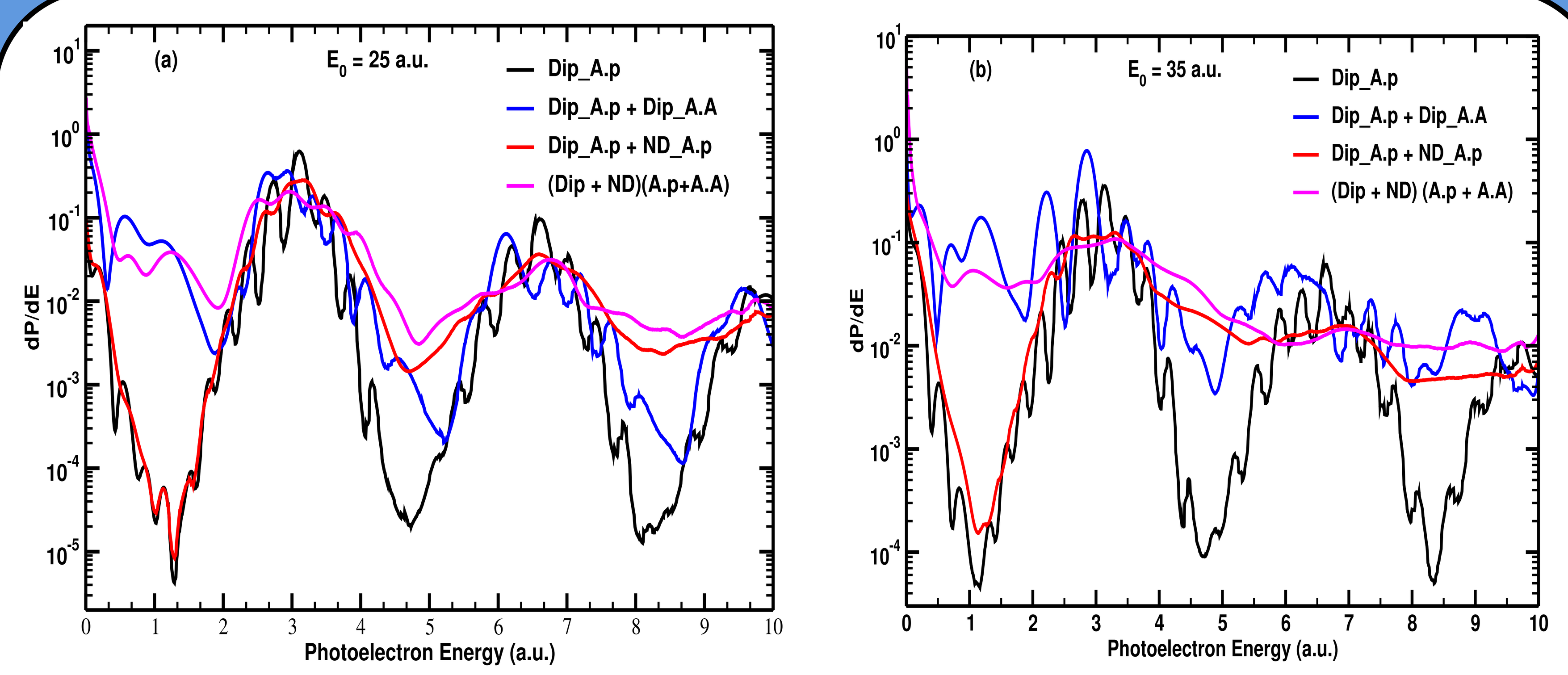

Figure 3. PE spectra at a peak electric field strength of (a)25 au (b)35 au for 15 cycle 13 nm laser pulse. Comparison of relative contributions of the multipole $\mathbf{A}\cdot\mathbf{p}$ and $\mathbf{A}^2$ components up to the quadrupole term. Black: dipole $\mathbf{A}\cdot\mathbf{p}$, Blue: dipole $\mathbf{A}\cdot\mathbf{p}$ plus dipole $\mathbf{A}^2$, Red: dipole plus quadrupole $\mathbf{A}\cdot\mathbf{p}$, and magenta: dipole plus quadrupole $\mathbf{A}\cdot\mathbf{p}+\mathbf{A}^2$ terms.

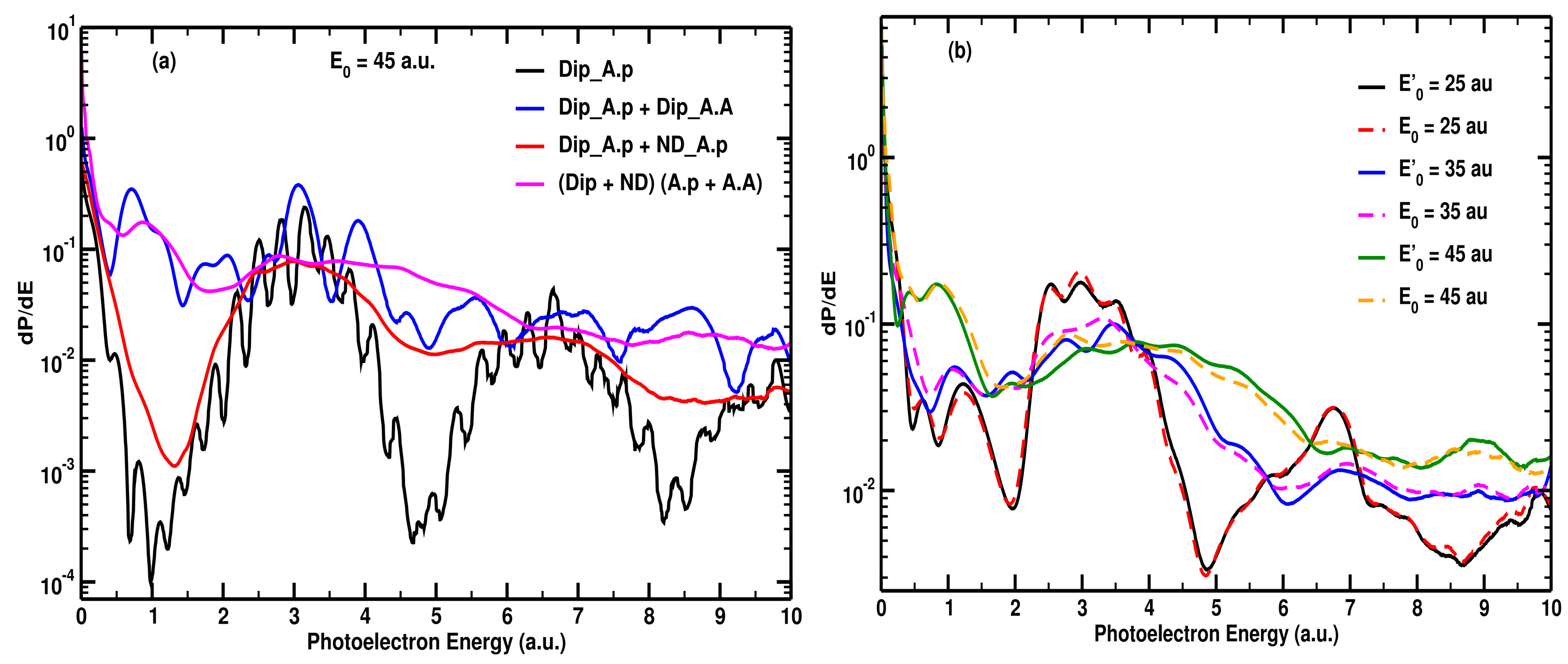

Figure 4. (a) Same as figure 3 but for a peak electric field strength of 45 au. (b) Multipole PE spectra for the electric field amplitudes ($E_0 = 25, 35, 45$) considered. Solid lines: ignore the derivative part of the CEP in the electric field. Broken lines: includes the derivative part of the CEP. See $A_2(t)$ in the theory section.

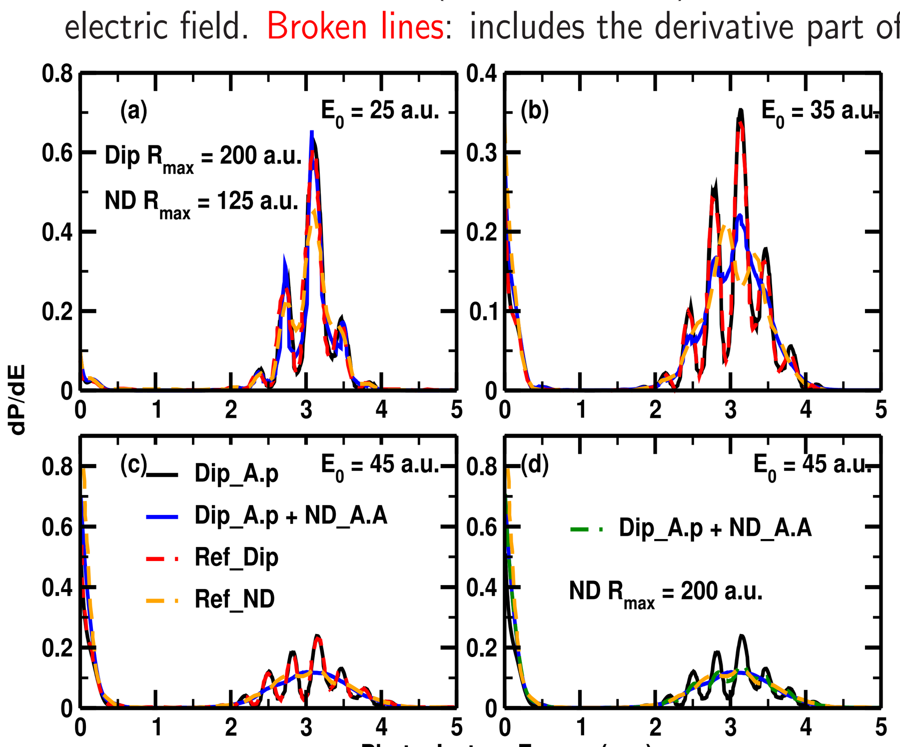

Figure 5. Comparison of the present 13 nm dipole $\mathbf{A}\cdot\mathbf{p}$ and the dipole plus quadrupole A^2 term with reference [2] results. The reference underestimates the impact of non-dipole effects.

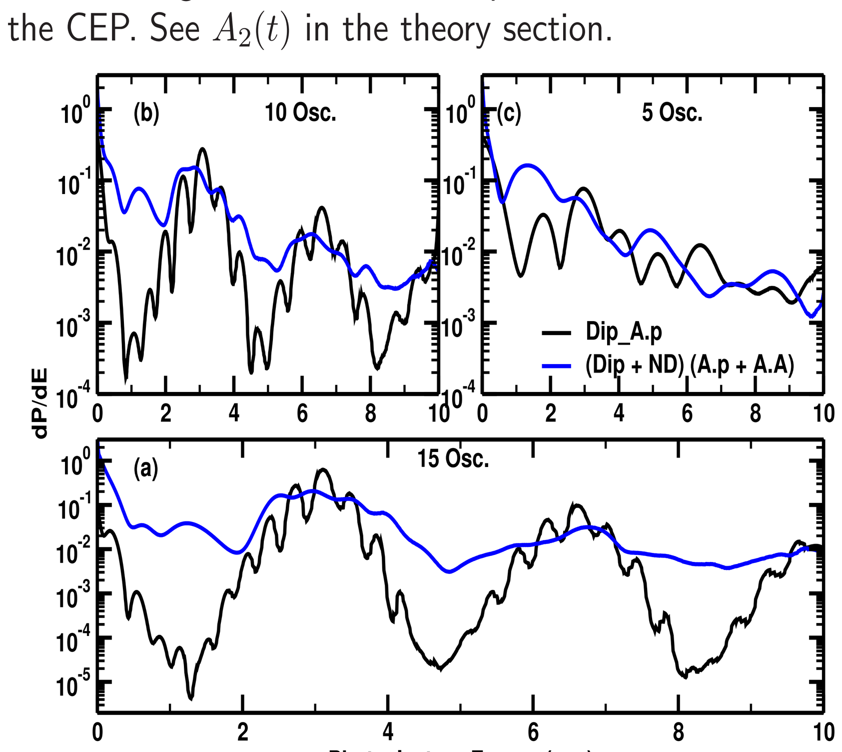

Figure 6. Variation of the present 13 nm dipole $\mathbf{A}\cdot\mathbf{p}$ and the dipole plus quadrupole $\mathbf{A}\cdot\mathbf{p}+A^2$ PE spectra with pulse duration for $E_0 = 25$ au. The non-dipole effects still quite significant even for short cycle pulses.

Conclusion

It is observed that:

- ponderomotive shift enhances (very) low energy structures energy spectrum at higher intensities .
- both $\mathbf{A}\cdot\mathbf{p}$ and A^2 terms have comparable beyond dipole corrections as long as U_p is not ignored.
- as intensity increases the non-dipole effects distort the ATI structures.
- non-dipole effects persist even in the short pulse limit though in this limit the beating pattern becomes visible.
- the published results of reference [2] were in excellent agreement with our dipole spectrum and quite a good agreement in the non-dipole spectrum for the interactions compared.

References

1. R. R. Freeman and P. H. Bucksbaum, *Investigations of above-threshold ionization using subpicosecond laser pulses*. Journal of Physics B: Atomic, Molecular and Optical Physics, Volume **24**, Number 2, p. 325, 1991.
2. M. Førre and A. S. Simonsen, *Nondipole ionization dynamics in atoms induced by intense xuv laser fields*. Phys. Rev. A **90**, 053411, (2014).
3. B. H. Bransden and C. J. Joachain, *Physics of atoms and molecules*. Longman Scientific and Technical, Essex, p.101, (1990).