Polychromatic resonant ionization of many-electron atoms

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Skobeltsyn Institute of Nuclear Physics
Lomonosov Moscow State University

1 August 2017, Cairns

The talk outline

The main goal is to investigate how quantum control of photoelectron angular distribution at bichromatic ionization is affected by structure of multi-electron atoms

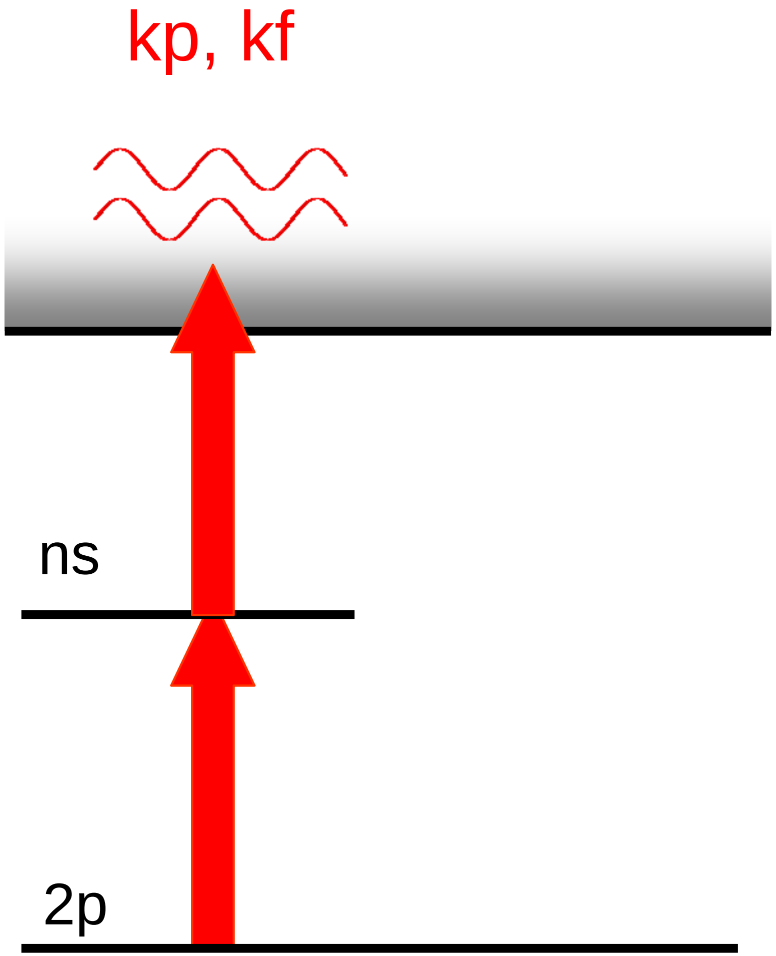
- The problem outline

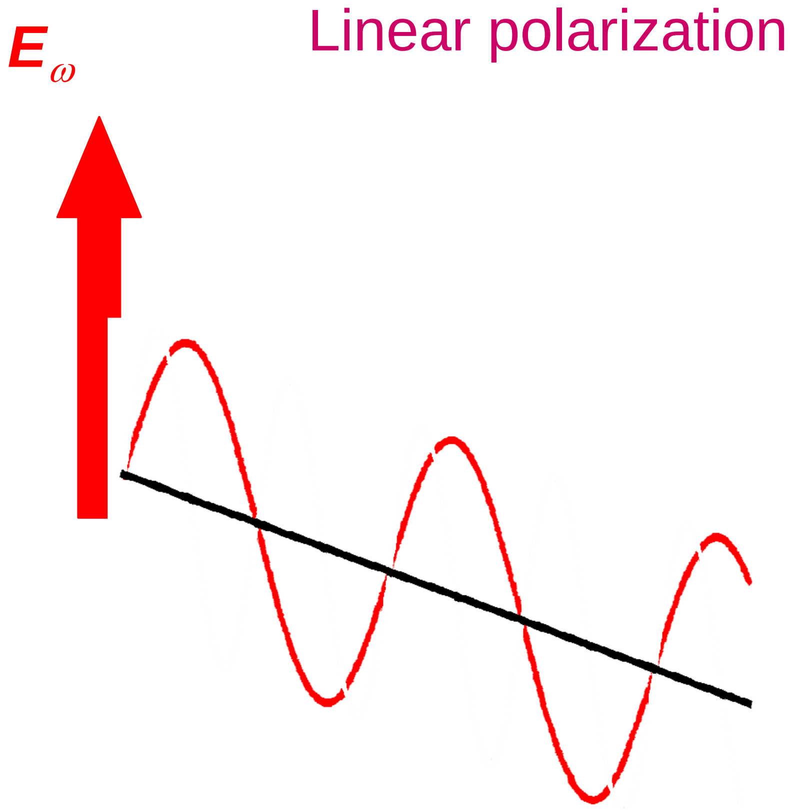
- Ionization of Neon by bichromatic field
  - with resonant excitation 3s state (single resonance)
  - with resonant excitation 4s state (double resonance)

- Role of temporal coherency and pulse intensity

- Concluding remarks

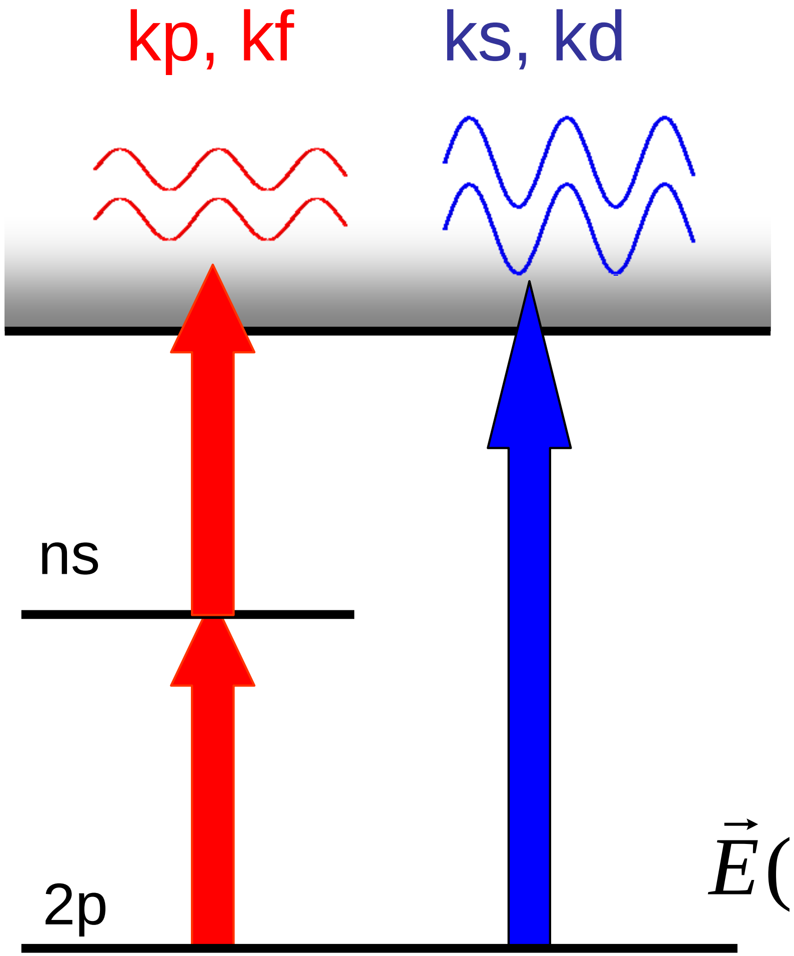
The scheme of bichromatic ionization

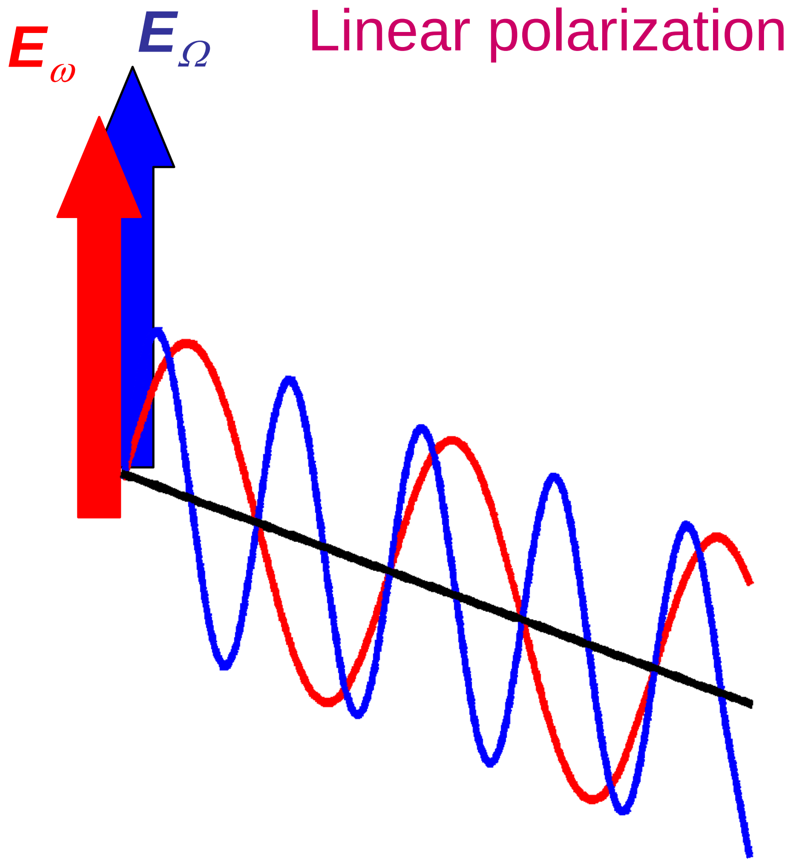




$$\vec{E}(t) = \vec{\varepsilon}\ E_0(t)\cos\omega t$$

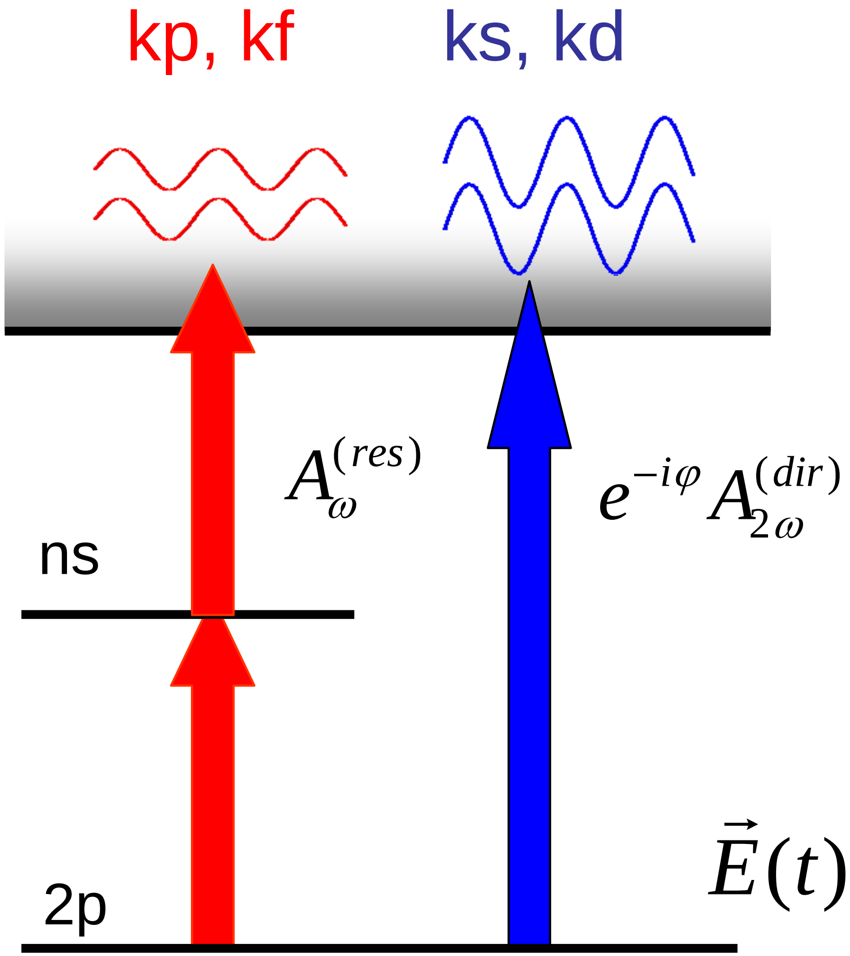
The scheme of bichromatic ionization

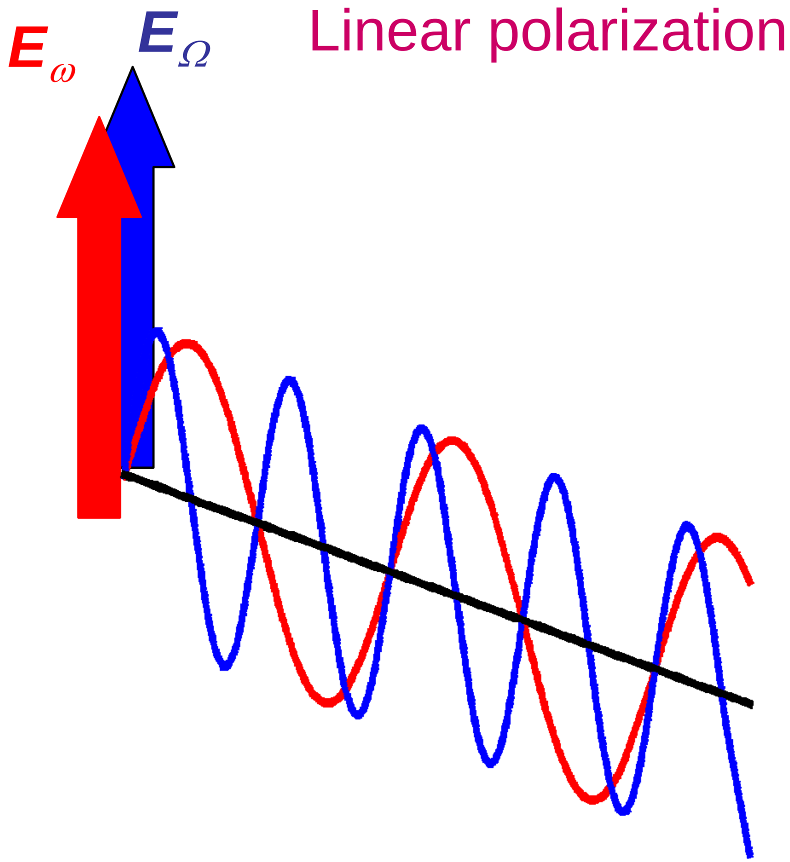




$$\vec{E}(t) = \vec{\varepsilon}\; E_0(t)\left(\cos\omega t + \eta\cos(2\omega t + \varphi)\right)$$

The scheme of bichromatic ionization

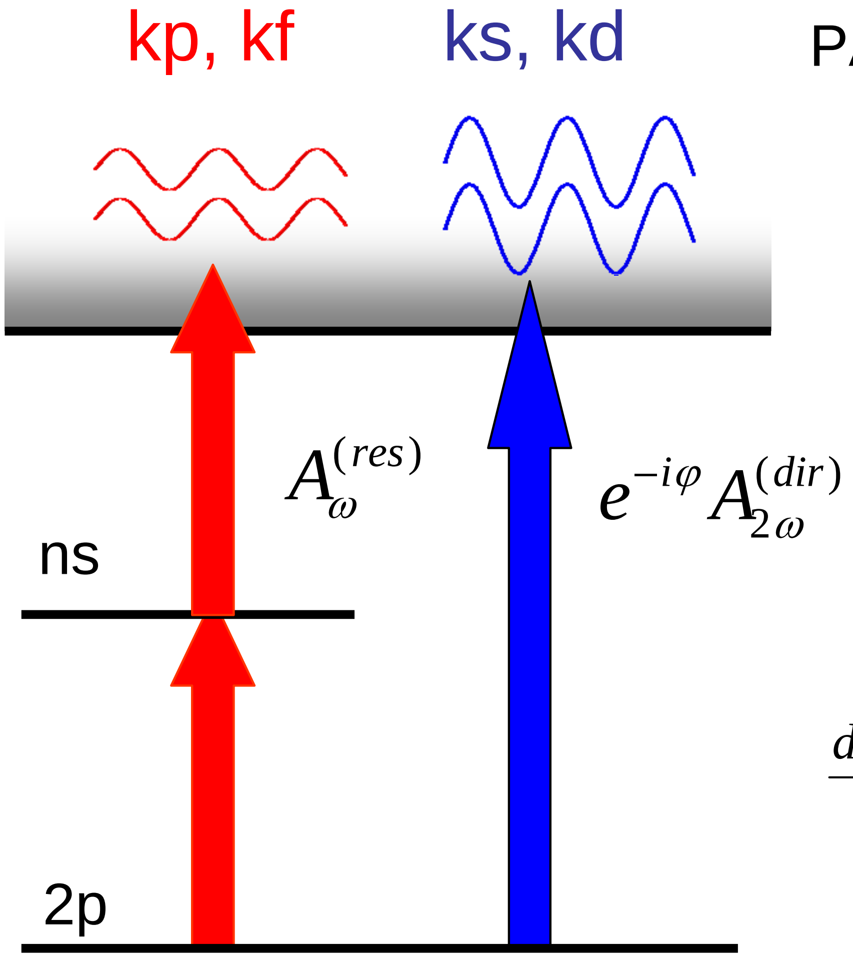


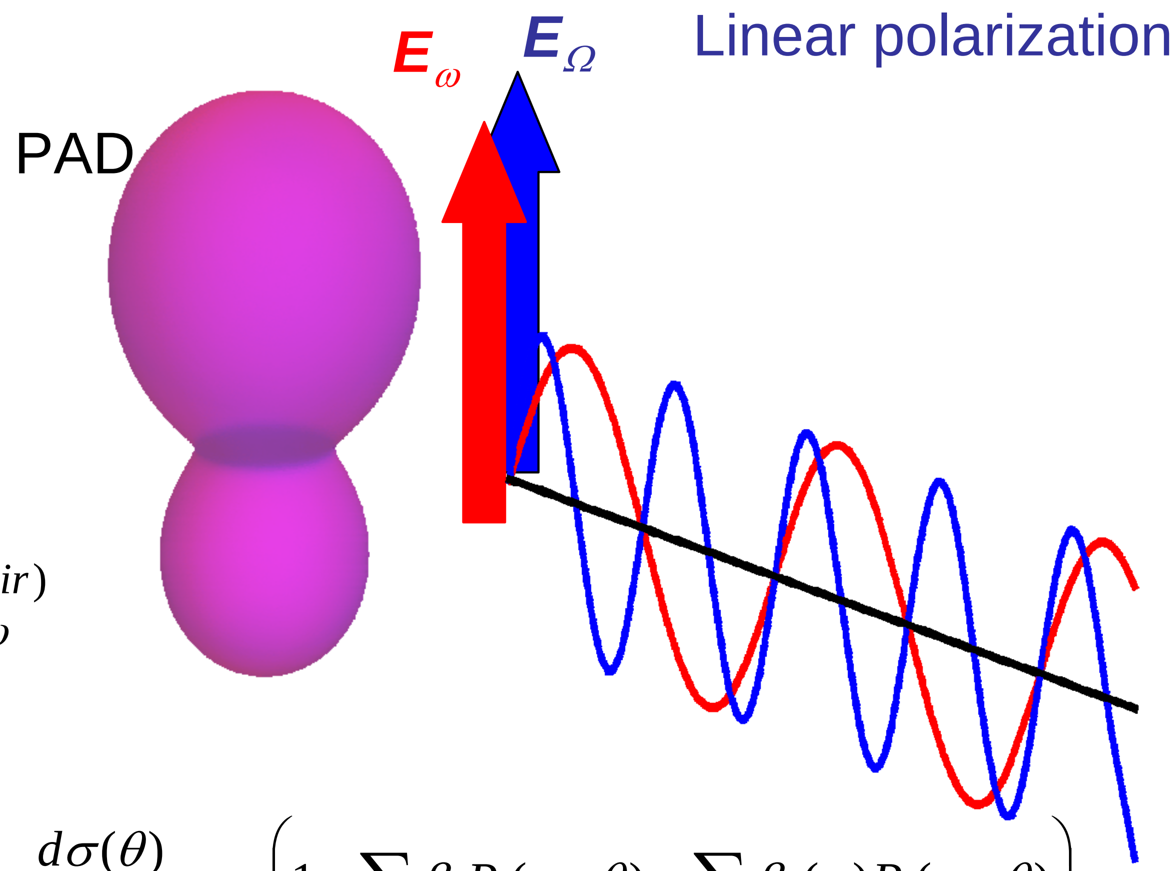


$$\vec{E}(t) = \vec{\varepsilon}\; E_0(t) \left(\cos \omega t + \eta \cos(2\omega t + \varphi)\right)$$

$$\left| A_{\omega}^{(res)} + e^{-i\varphi} A_{2\omega}^{(dir)} \right|^2$$

PAD in bichromatic ionization



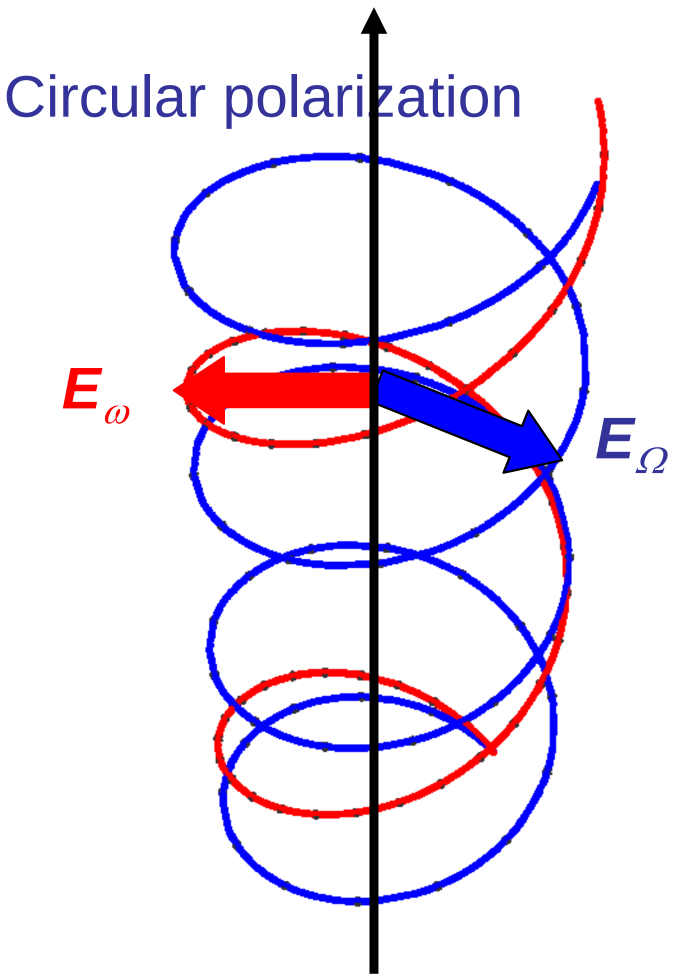


$$\frac{d\sigma(\theta)}{d\Omega} = \sigma_0\left(1 + \sum_{2,4}\beta_k P_k(\cos\theta) + \sum_{1,3}\beta_k(\varphi)P_k(\cos\theta)\right)$$

$$\left|A_{\omega}^{(res)} + e^{-i\varphi}A_{2\omega}^{(dir)}\right|^2$$

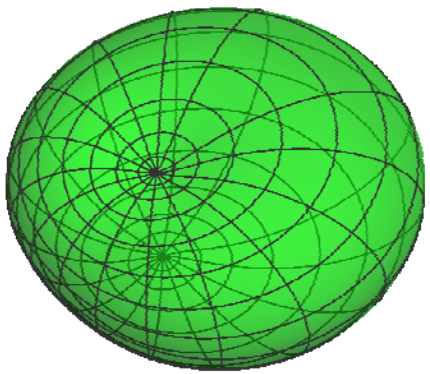
The interference of pathways with different parities leads to PAD asymmetry

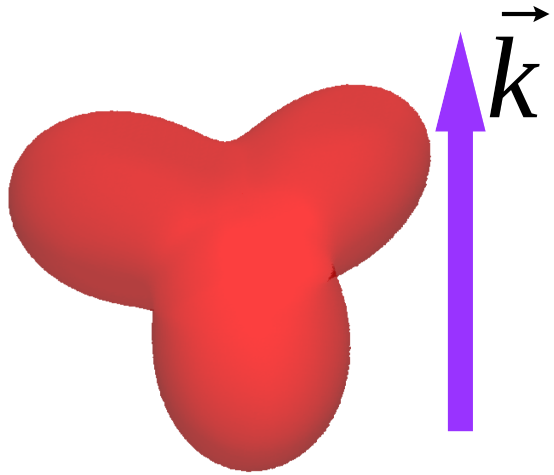
PAD in bichromatic ionization



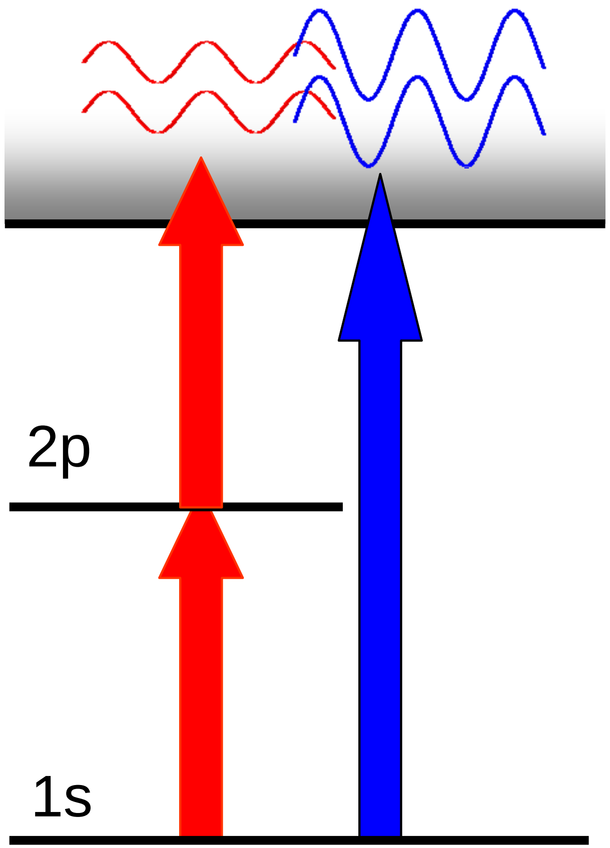
$$\frac{d\sigma(\theta)}{d\Omega} = \sigma_0\left(1 + \sum_{2,4}\beta_k P_k(\cos\theta) + \left|\gamma_{3\mu}\right|\sin^3\theta\cos(\mu\varphi + \psi)\right)$$

PAD

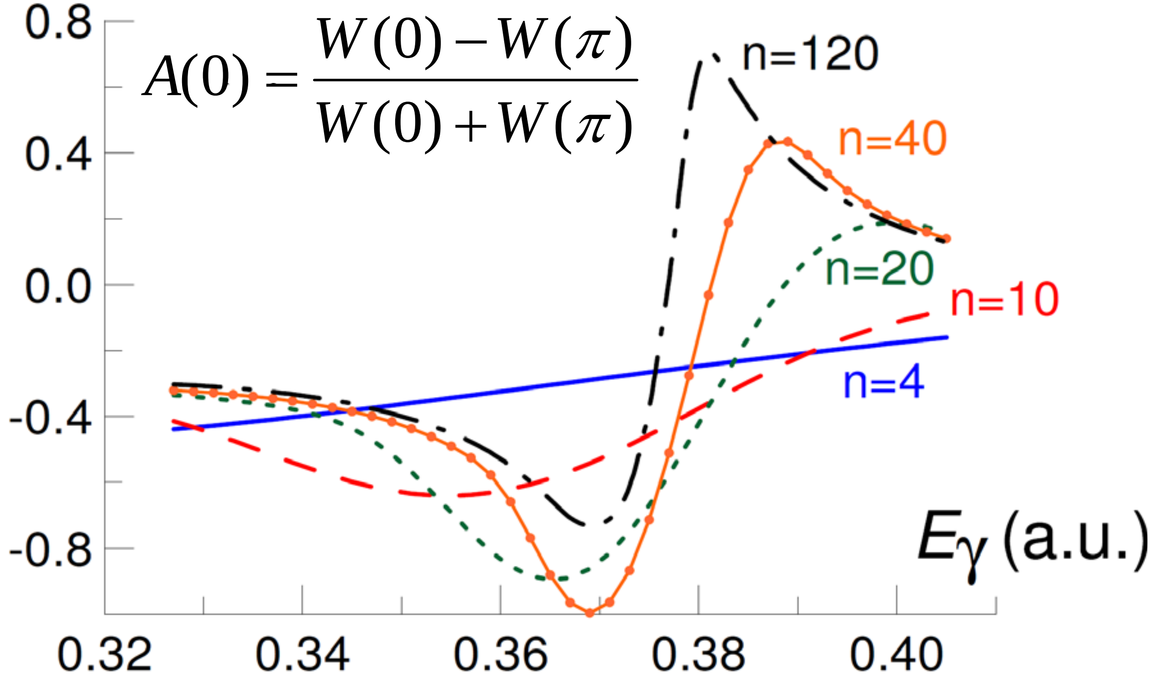




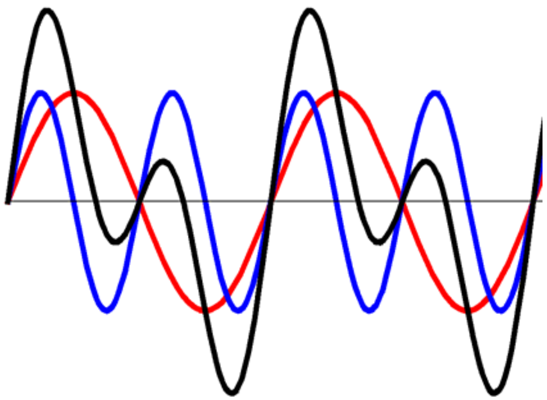
Formation of the asymmetry



hydrogen



N is number of optical cycles



n=10 ~ 4 fs

The main features of (ω+2ω) coherent control

✓The effect is observed in differential parameters (β and/or asymmetry of PAD)

✓Exists for infinite pulse and actually takes time to form

✓It is independent of absolute value of initial phase

✓Exists in perturbative regime

The experiment

LETTERS
PUBLISHED ONLINE: 22 FEBRUARY 2016 | DOI: 10.1038/NPHOTON.2016.13
nature photonics

Coherent control with a short-wavelength free-electron laser.

Nature Photonics **10**,176, (2016).

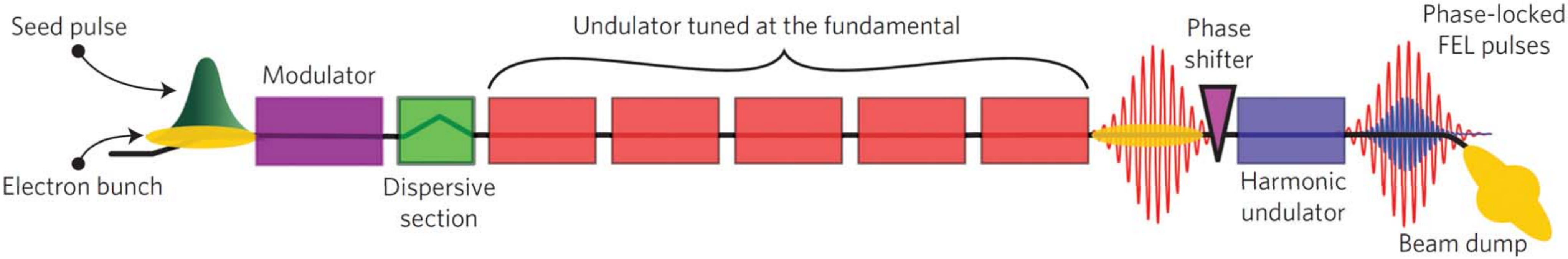
K. C. Prince, E. Allaria, C. Callegari, R. Cucini, G. De Ninno, S. Di Mitri, B. Diviacco,E. Ferrari, P. Finetti, D. Gauthier, L. Giannessi, N. Mahne, G. Penco, O. Plekan, L. Raimondi, P. Rebernik, E. Roussel, C. Svetina, M. Trovò, M. Zangrando, M. Negro, P. Carpeggiani, M. Reduzzi, G. Sansone, A. N. Grum-Grzhimailo, E. V. Gryzlova, S. I. Strakhova, K. Bartschat, N. Douguet, J. Venzke, D. Iablonskyi, Y. Kumagai, T. Takanashi, K. Ueda, A. Fischer, M. Coreno, F. Stienkemeier, Y. Ovcharenko, T. Mazza and M. Meyer

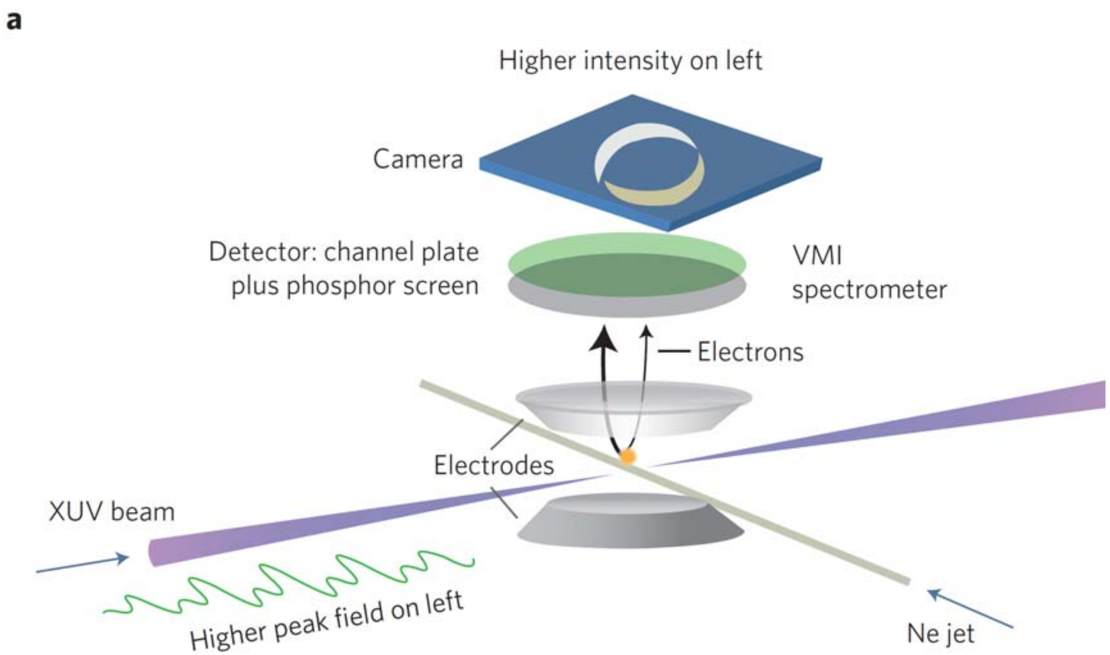
Interfering one-photon and two-photon ionization byfemtosecond VUV pulses in the region of an intermediate resonance

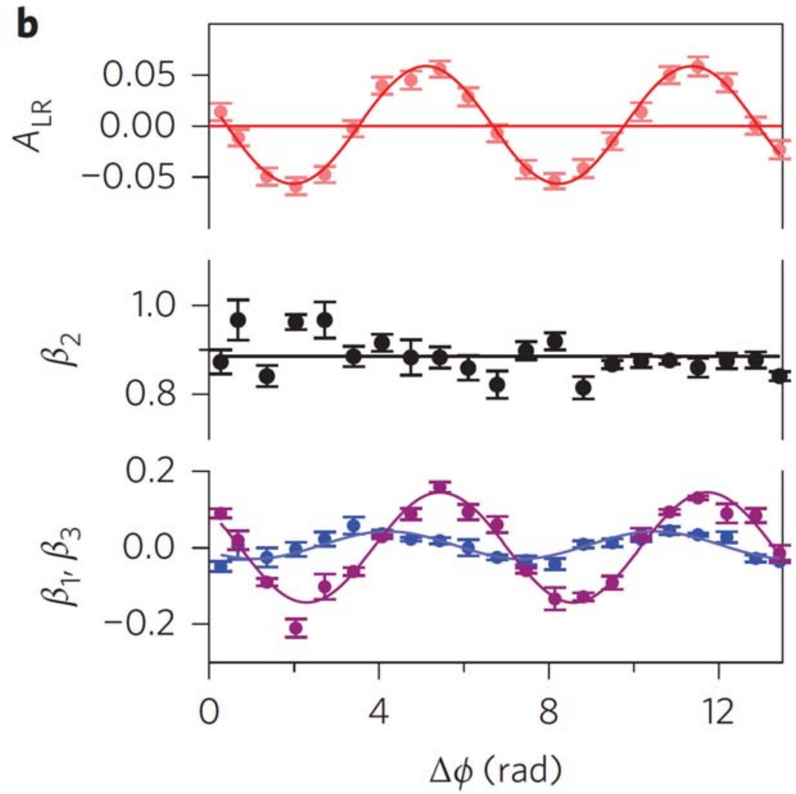
. *Phys. Rev. A* **91**, 063418, (2015).

A.N. Grum-Grzhimailo, E.V. Gryzlova, E. I. Staroselskaya, J. Venzke, and K. Bartschat

The experiment







Theoretical approaches

✓**Non-stationary perturbation theory** (includes fine-structure effects)
Allow to get suitable parameterization of PAD; analyze role of particular amplitudes (LSJ)

✓**TDSE** (single active electron approach and model potential. [A. N. Grum-Grzhimailo et al J. Phys. B **39**, 4659 (2006)], to analyze strong field effects, state depletion and to check applicability of PT approach) (TDSE)

✓**Stationary perturbation theory**
Allow to find analytical dependency on pulse parameter (LS-inf)

Moscow team
Alexei N. Grum-Grzhimailo, Elena V. Gryzlova, Ekaterina I. Staroselskaya,
Deake team
Nicolas Douguet and Klaus Bartschat

Pulse parameters

$$\vec{E}(t) = \vec{\varepsilon}\, E_0(t) \left(\cos\omega t + \eta \cos(2\omega t + \varphi)\right)$$

polarization

envelope

Pulse duration

Intensity

Form

Ratio of first and second harmonics

Relative phase

Observable parameters

$$\frac{d\sigma(\theta)}{d\Omega} = \sigma_0 \left(1 + \sum_{2,4} \beta_k P_k(\cos\theta) + \sum_{1,3} \beta_k(\varphi) P_k(\cos\theta) \right)$$

Incoherent sum of single- and two-photon amplitudes

Interference of single- and two-photon amplitudes

Asymmetry

$$A(0) = \frac{\beta_1 + \beta_3}{1 + \beta_2 + \beta_4}$$

Odd parameters depend on phase between harmonics as cosine, so it is instructive to compare maximal amplitude and corresponding phase $A(0) = |A| \cos(\varphi - \varphi_m)$

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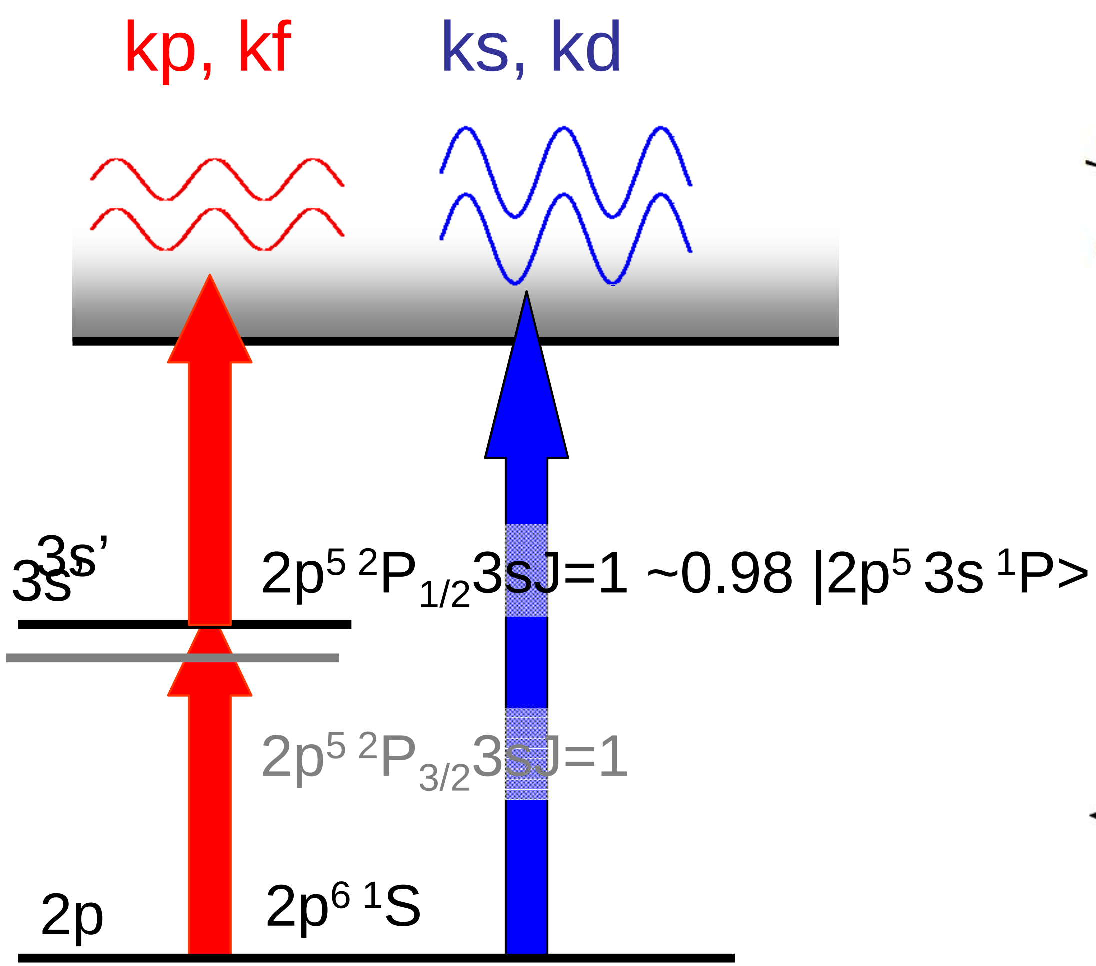
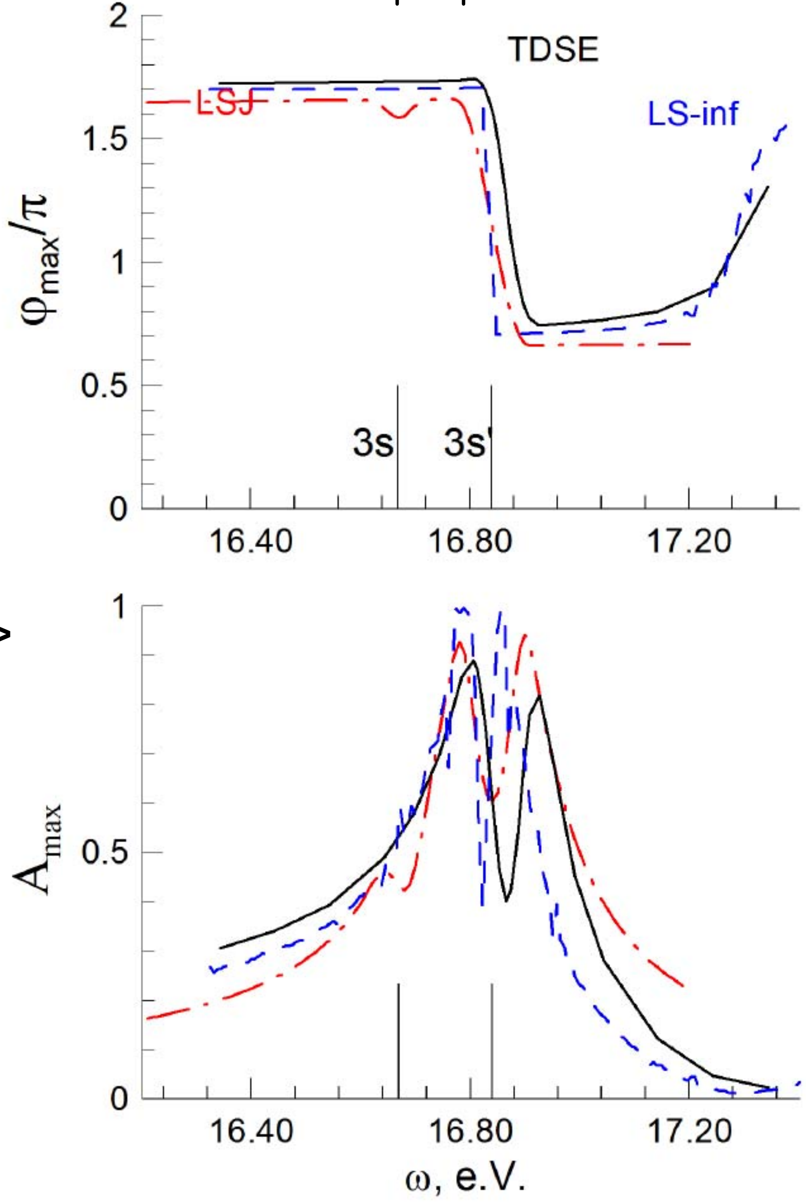
Ionization of Neon in the vicinity of $2p^5\,3s\;^1P$



$$A(0) = |A|\cos(\varphi - \varphi_m)$$



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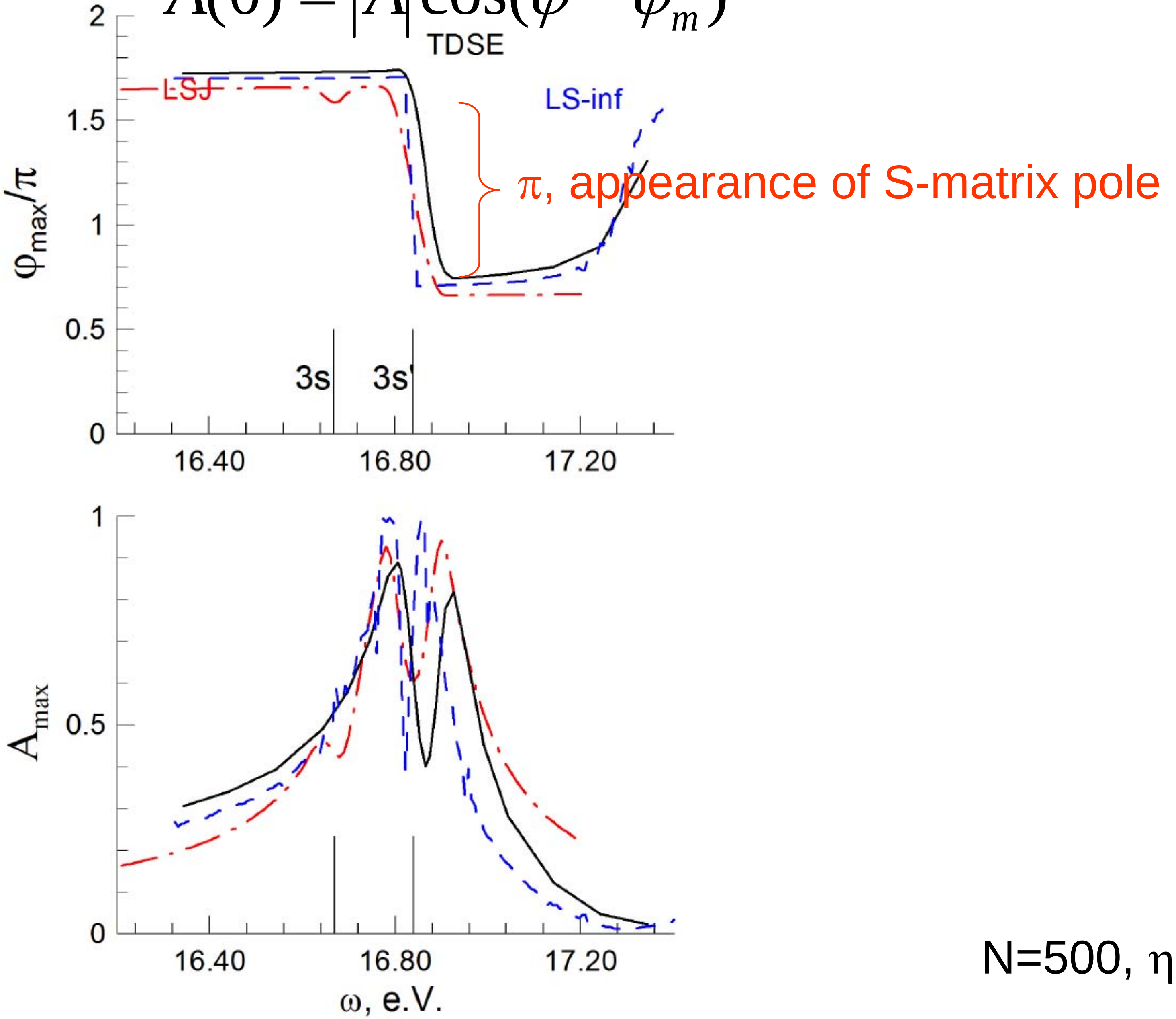
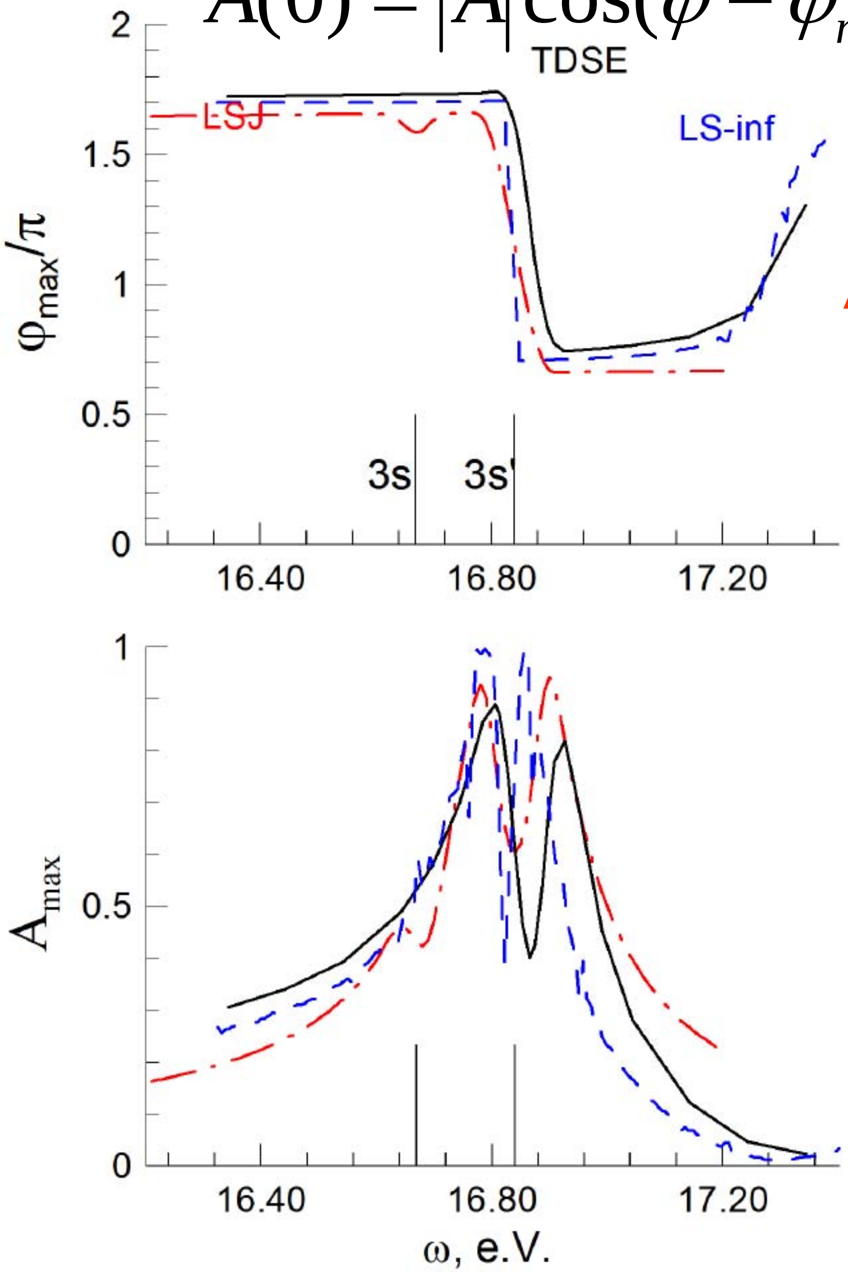


N=500, η=0.1, I=10^{12} W/cm^2

Ionization of Neon in the vicinity of $2p^5\,3s\,^1P$

$$A(0) = |A| \cos(\varphi - \varphi_m)$$

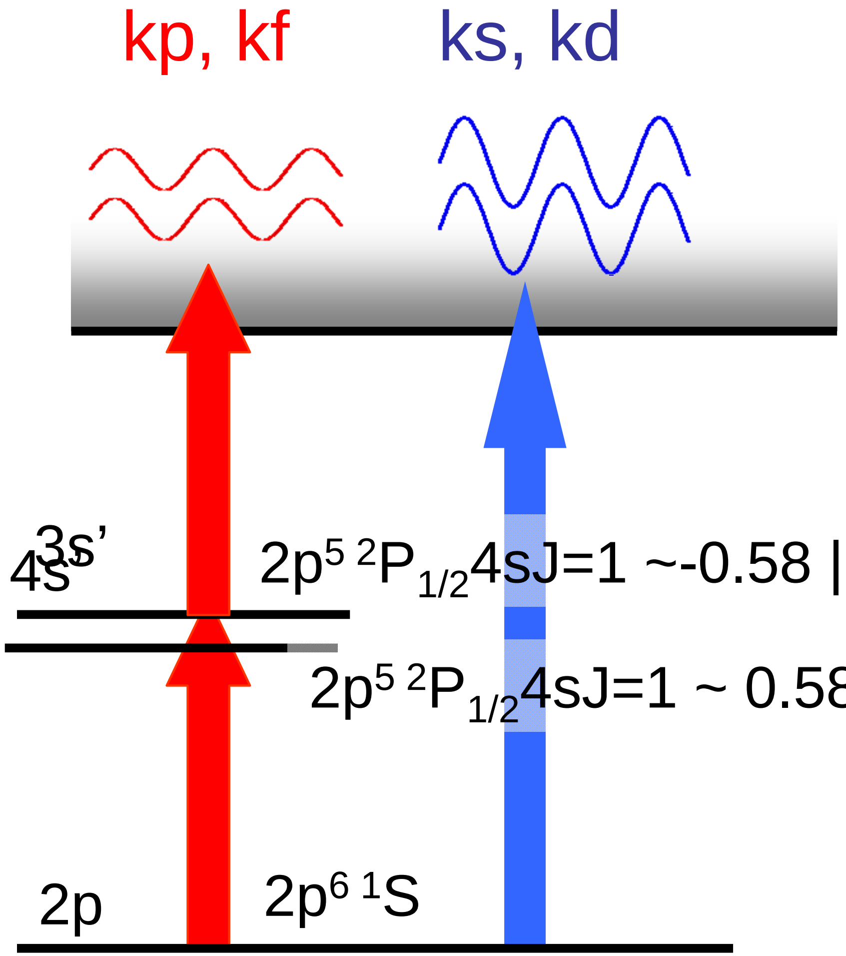


Asymmetry pass through zero

N=500, η=0.1, I=10^{12} W/cm^2

Ionization of Neon in the vicinity of $2p^5\,4s\,^1P$

$$\hat{J}_f\hat{S}\begin{Bmatrix} S & L & J \\ J_f & j & S_f \end{Bmatrix} \begin{matrix} \approx -\sqrt{\dfrac{2}{3}}\left|{}^1P\right\rangle + \dfrac{1}{\sqrt{3}}\left|{}^3P\right\rangle \\ \approx \sqrt{\dfrac{2}{3}}\left|{}^3P\right\rangle + \dfrac{1}{\sqrt{3}}\left|{}^1P\right\rangle \end{matrix}$$

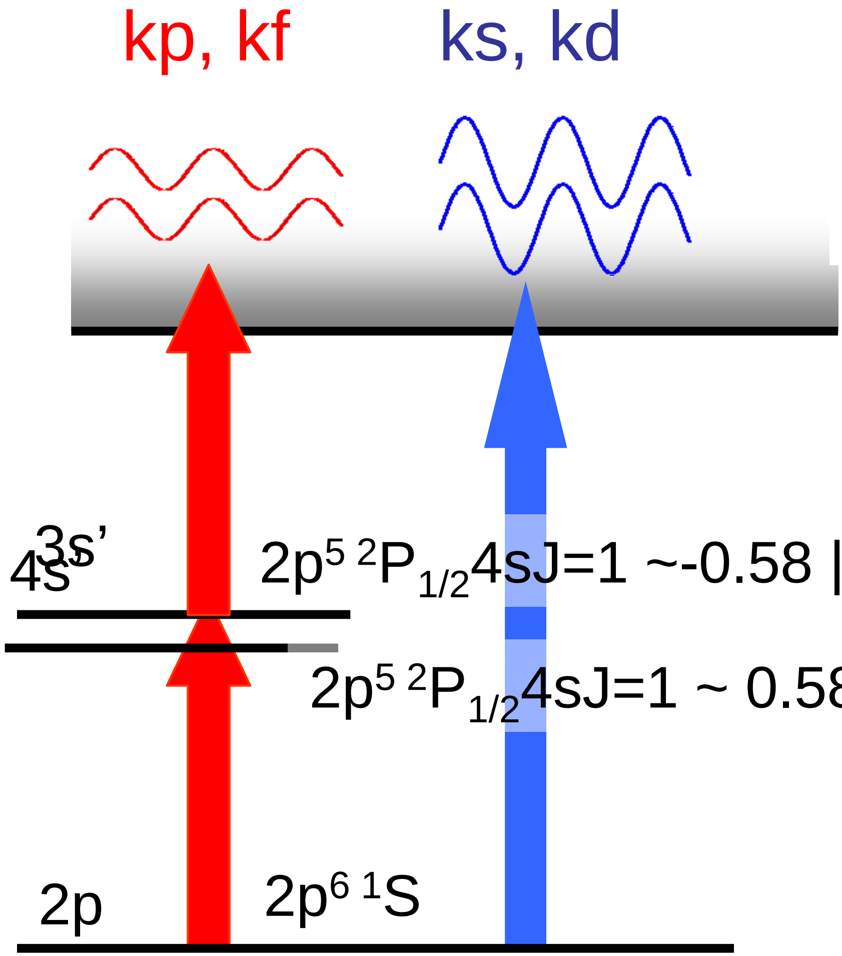


$2p^5\,^2P_{1/2}4sJ=1 \sim -0.58\ |2p^5\,4s\,^3P\rangle + 0.81|2p^5\,4s\,^1P\rangle$

$2p^5\,^2P_{1/2}4sJ=1 \sim 0.58\ |2p^5\,4s\,^1P\rangle + 0.81|2p^5\,4s\,^3P\rangle$

Ionization of Neon in the vicinity of $2p^5\,4s\,^1P$

Interference is possible only for S=S'



$$\frac{dW^{(III)}}{d\Omega} = \eta \frac{(-1)^{L_f}}{2\pi\sqrt{3}} \mathrm{Re}\left[\sum_{ll'L'k} \hat{l}\hat{l}'(l0,l'0\,|\,k0)(10,L'0\,|\,k0)(10,10\,|\,L'0)\begin{Bmatrix} l & l' & k\\ L' & 1 & L_f\end{Bmatrix}\right.$$
$$\left.\times e^{i(\Delta_l^{(1)}-\delta_{l'}^{(2)})} D^{(0)}_{0\to L_f l,1} T^{(1)} \left(\sum_n \left|\alpha_{10}^{\zeta_n}\right|^2 T^{(2)}_{E_n} D^{(0)*}_{n,1\to L_f l',L'} D^{(0)*}_{0\to n,1}\right)\right] P_k(\cos\vartheta)\,,$$

$2p^5\,^2P_{1/2}4sJ=1 \sim -0.58\,|2p^5\,4s\,^3P\rangle + 0.81|2p^5\,4s\,^1P\rangle$

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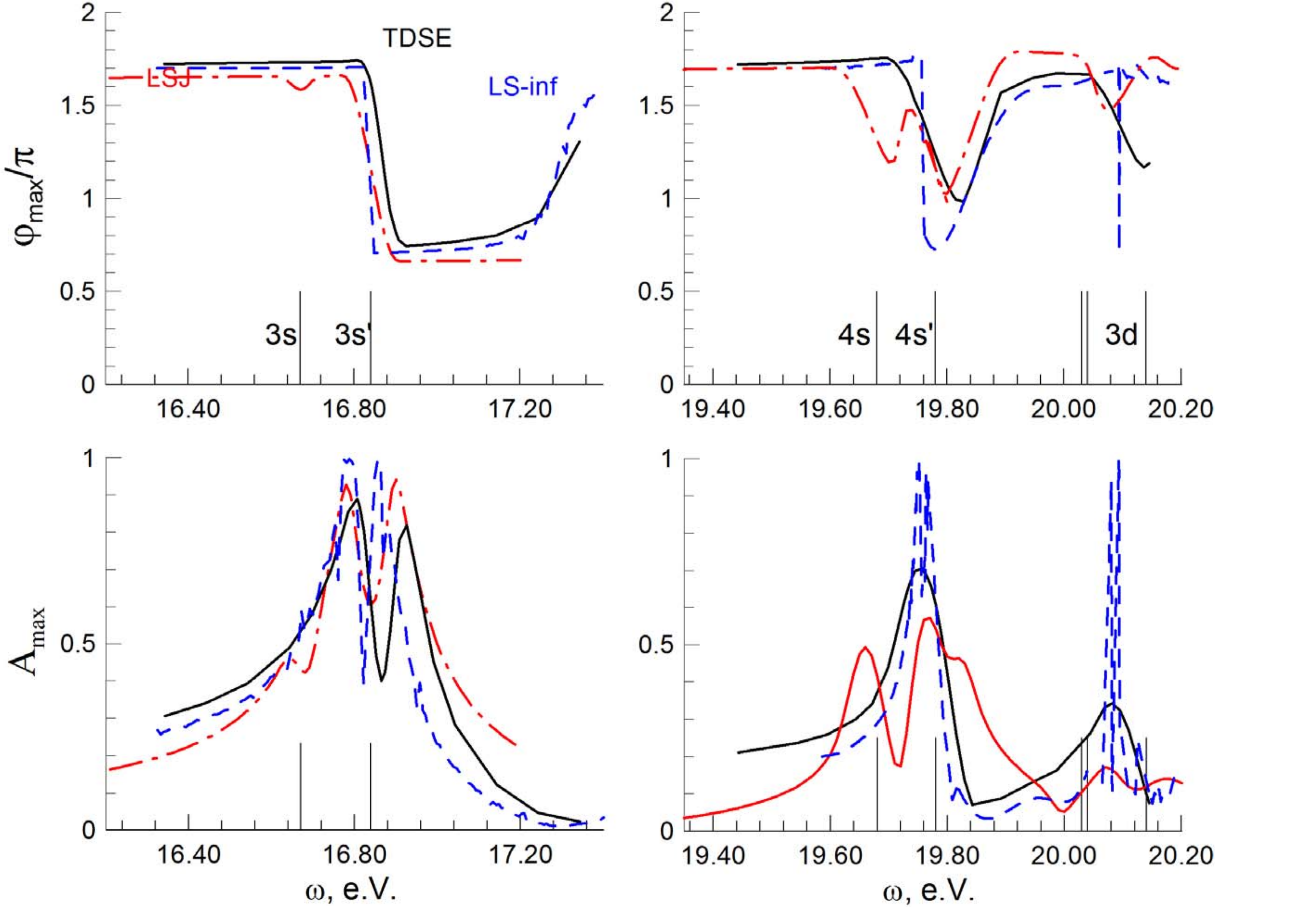
Ionization of Neon in the vicinity of $2p^5\ ns\ ^1P$



N=500, η=0.1, I=10^{12} W/cm^2

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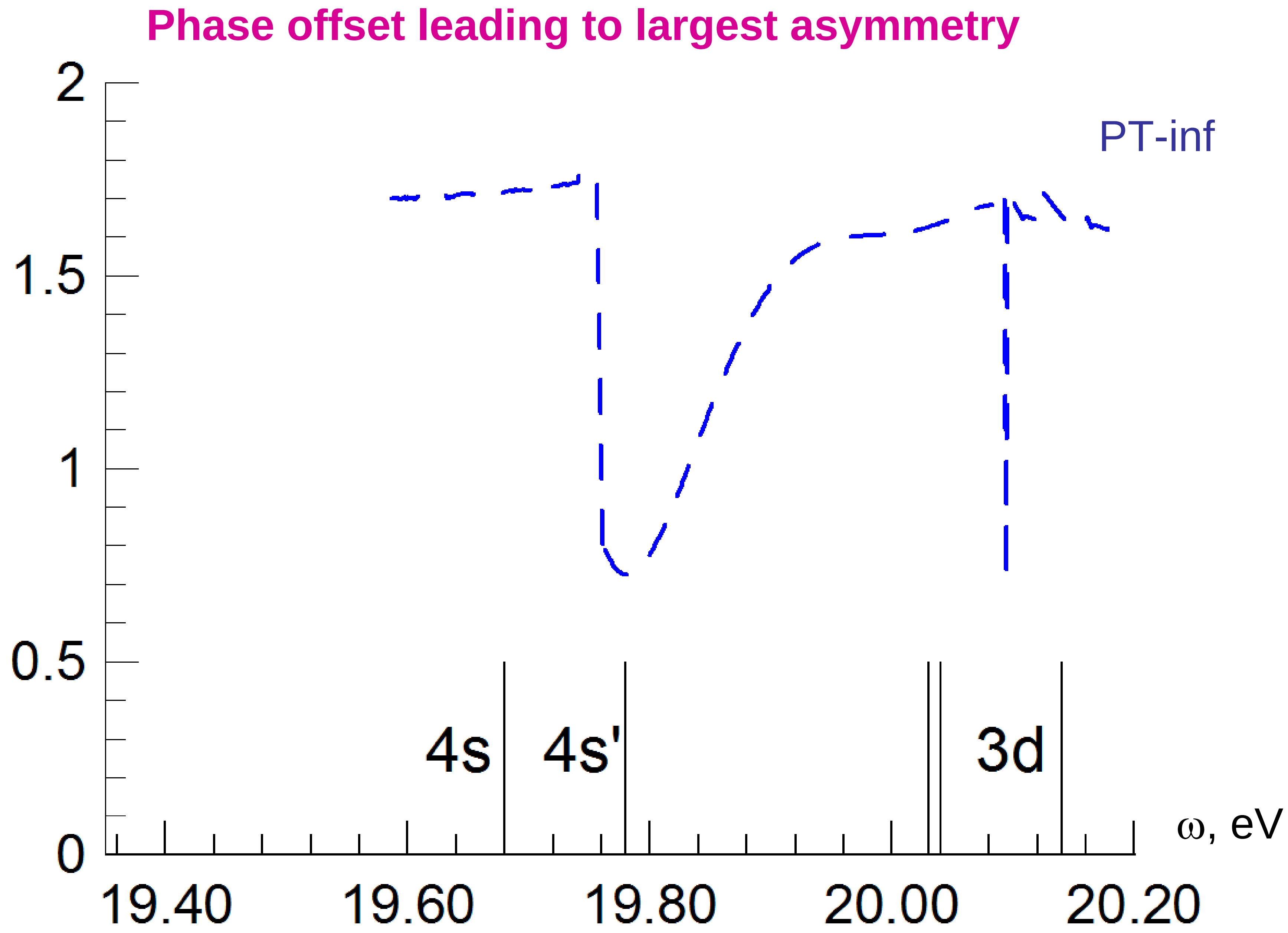

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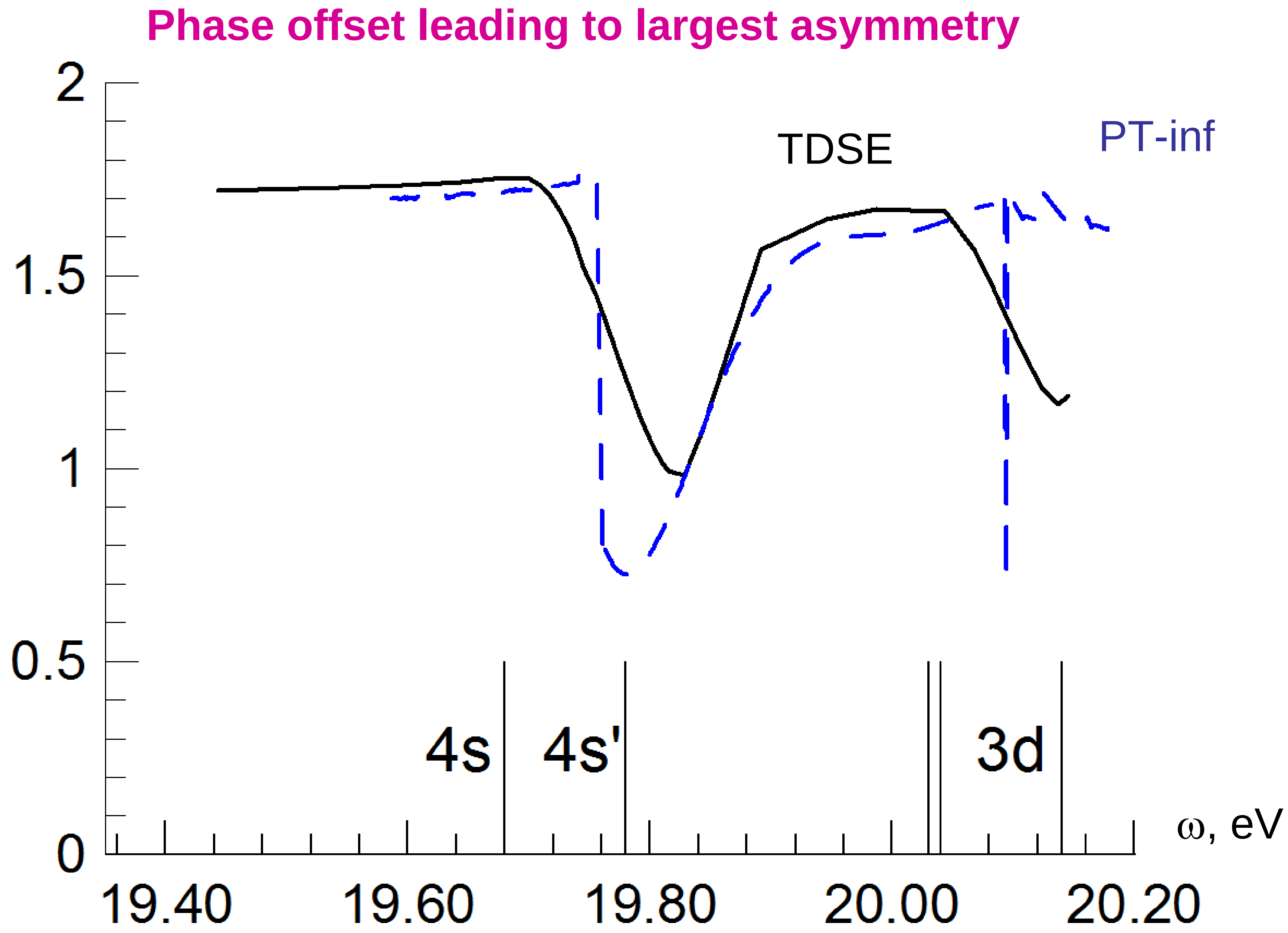

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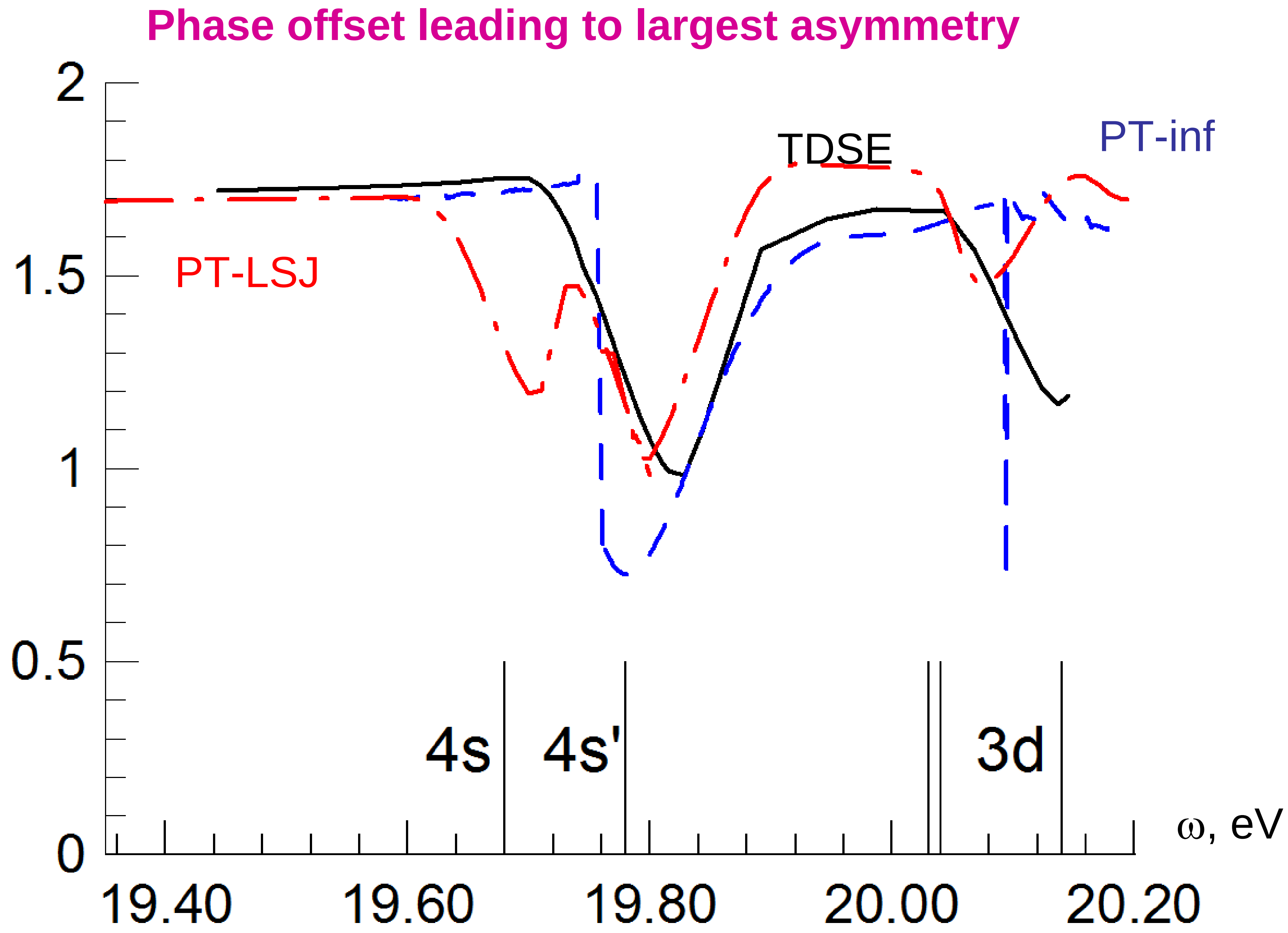
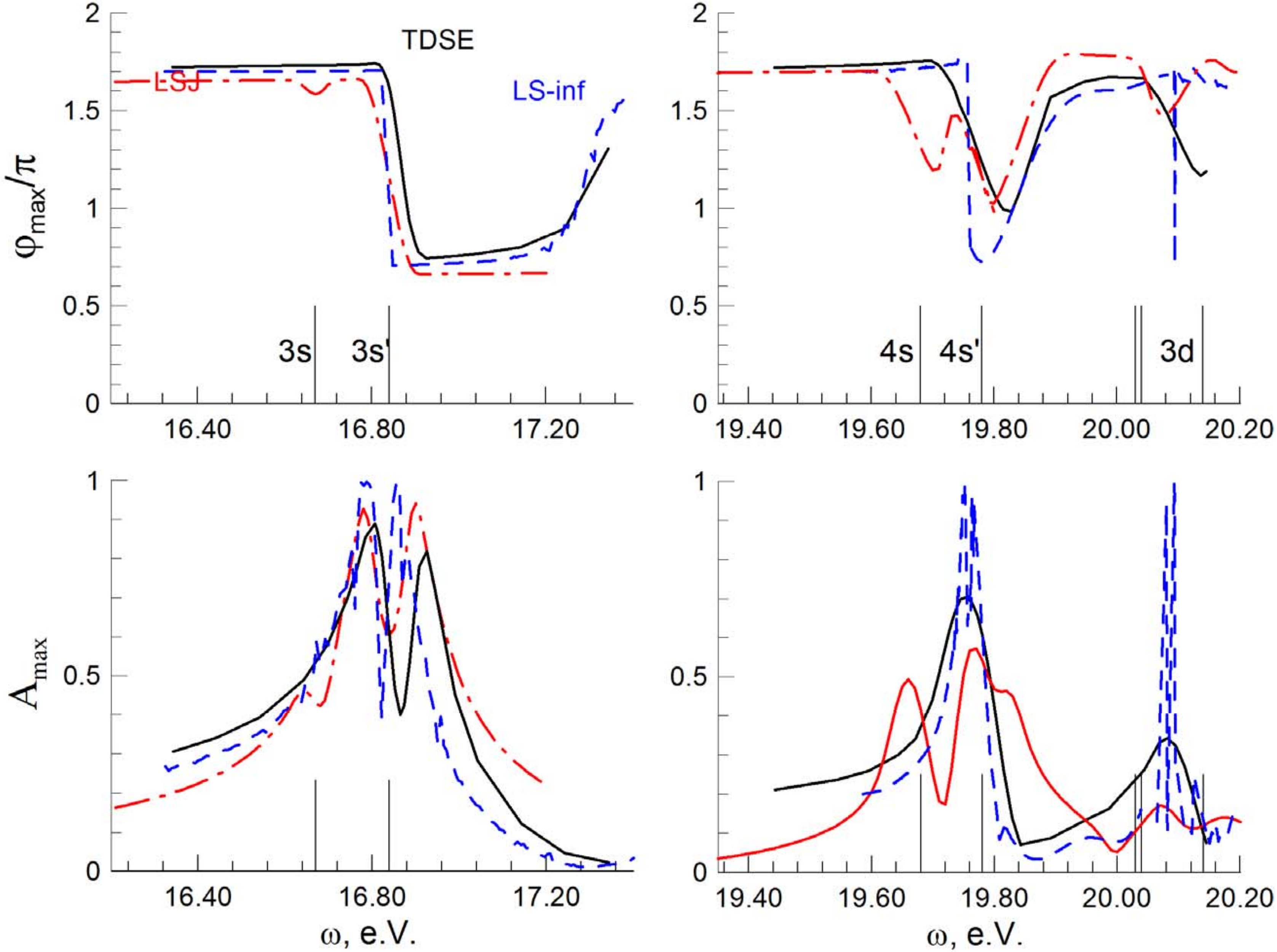


Ionization of Neon in the vicinity of $2p^5$ ns ^1P



N=500, η=0.1, I=10^{12} W/cm^2

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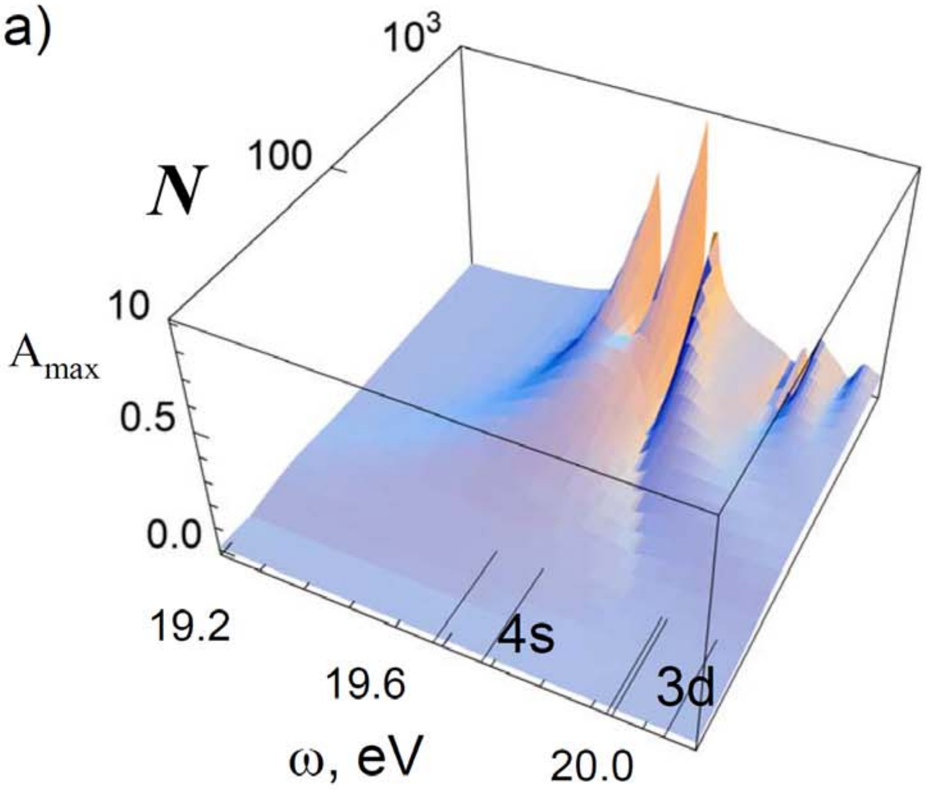
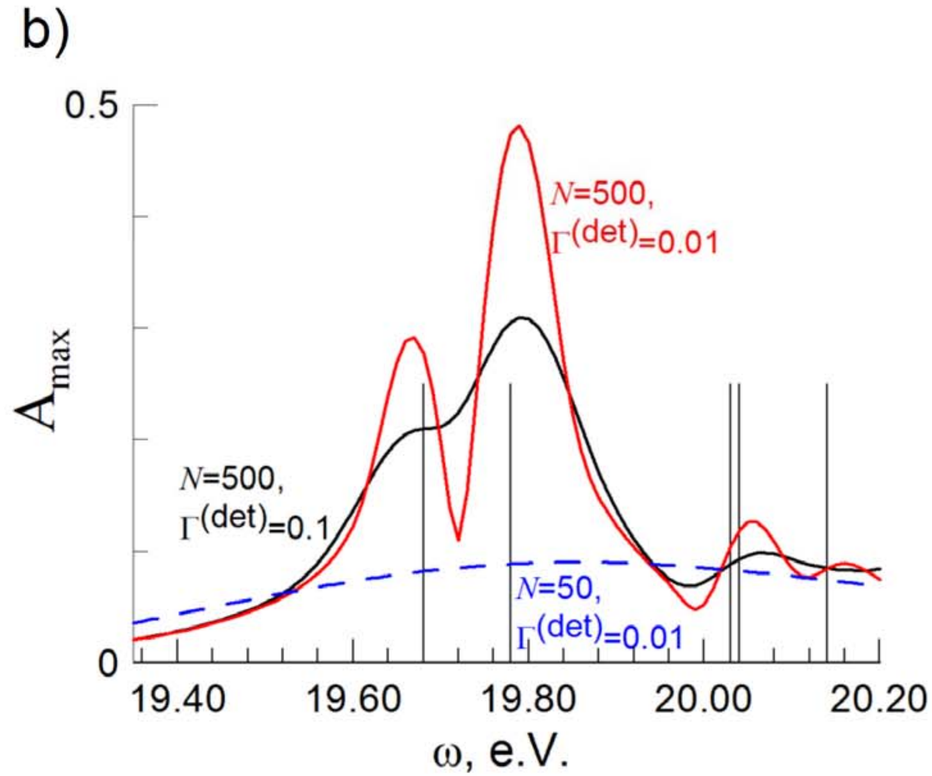
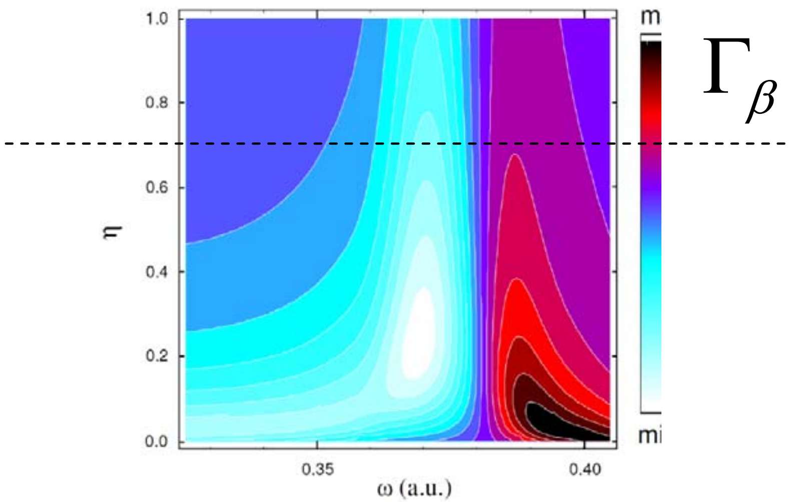
Effect of pulse coherency

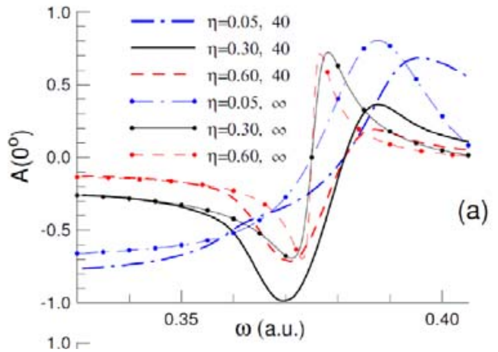




Harmonic ratio (η)



$$\Gamma_\beta = \Gamma_\gamma$$



Two-photon amplitude ~ *I*

Single photon amplitude $\eta\sqrt{I}$

$$\sqrt{I}/\eta = const$$

Leads to the same asymmetry

Fano-like profile

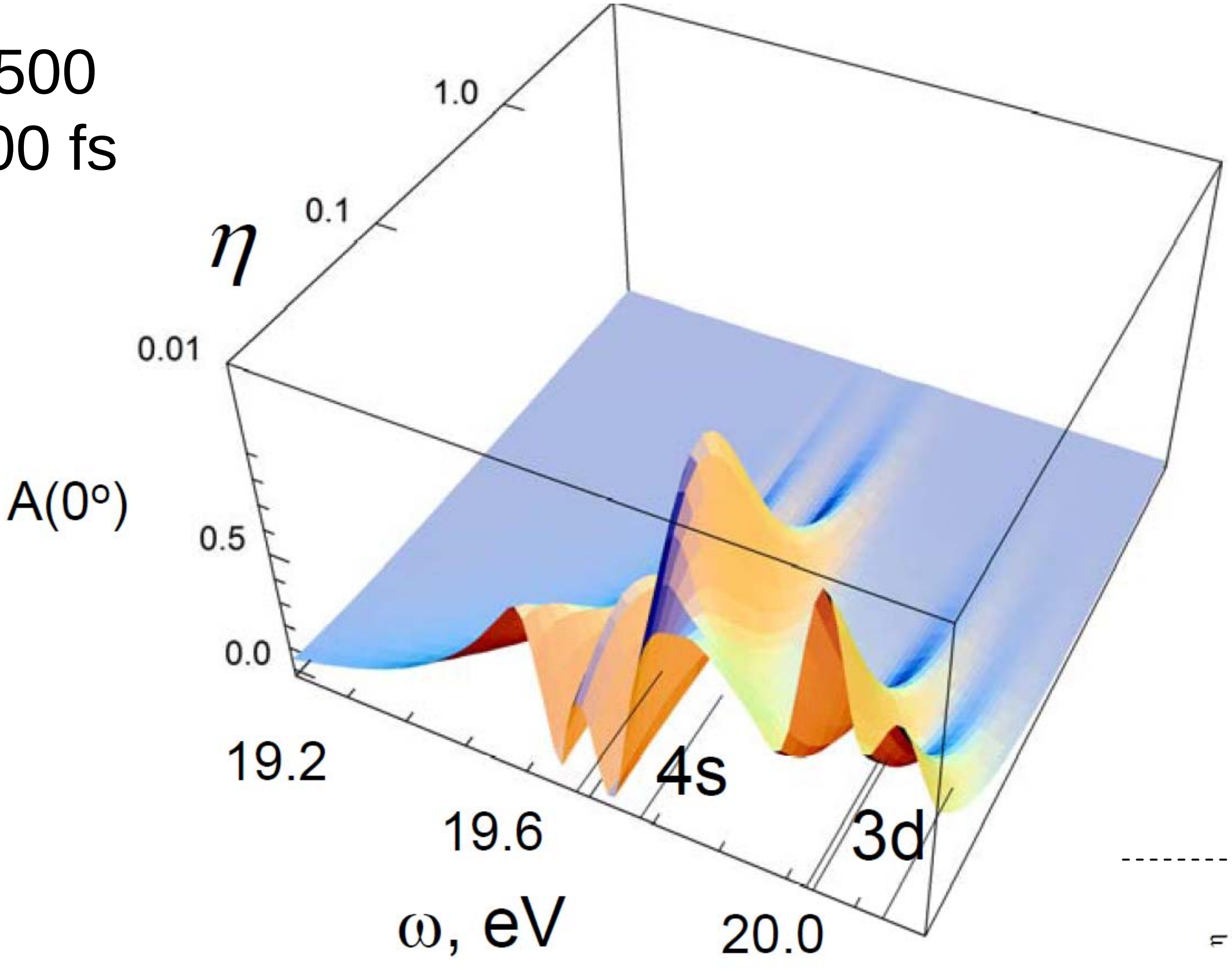
$$\beta_2 = 2\left(1 - \frac{B_2}{1+\varepsilon^2}\right); \qquad \beta_1 = \frac{B_1\varepsilon}{1+\varepsilon^2}\cos(\varphi - \varphi_1); \qquad \varepsilon = \frac{2\,\Delta\omega}{\Gamma_\beta};$$

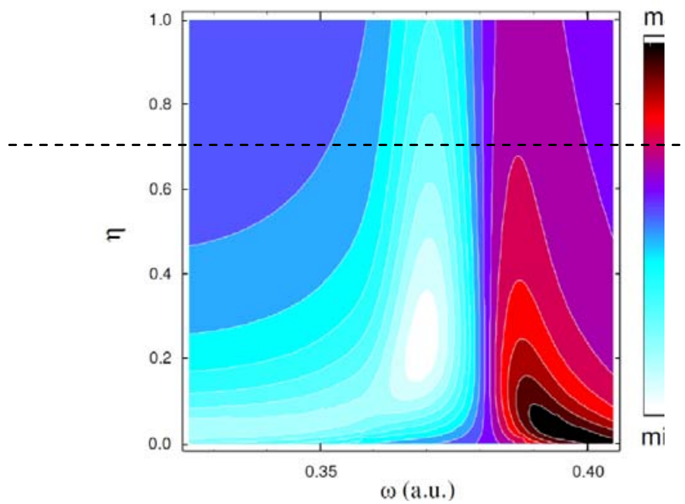
$$\beta_4 = \frac{B_4}{1+\varepsilon^2}; \qquad \beta_3 = \frac{B_3\varepsilon}{1+\varepsilon^2}\cos(\varphi - \varphi_3); \qquad \Gamma_\beta = \frac{\sqrt{I}}{2\eta}\cdot\frac{\sqrt{\left|D_s^{(2)}\right|^2 + \left|D_d^{(2)}\right|^2}}{\left|D_p^{(1)}\right|}$$

Phys. Rev. A **91**, 063418, (2015).

Role of harmonics ratio

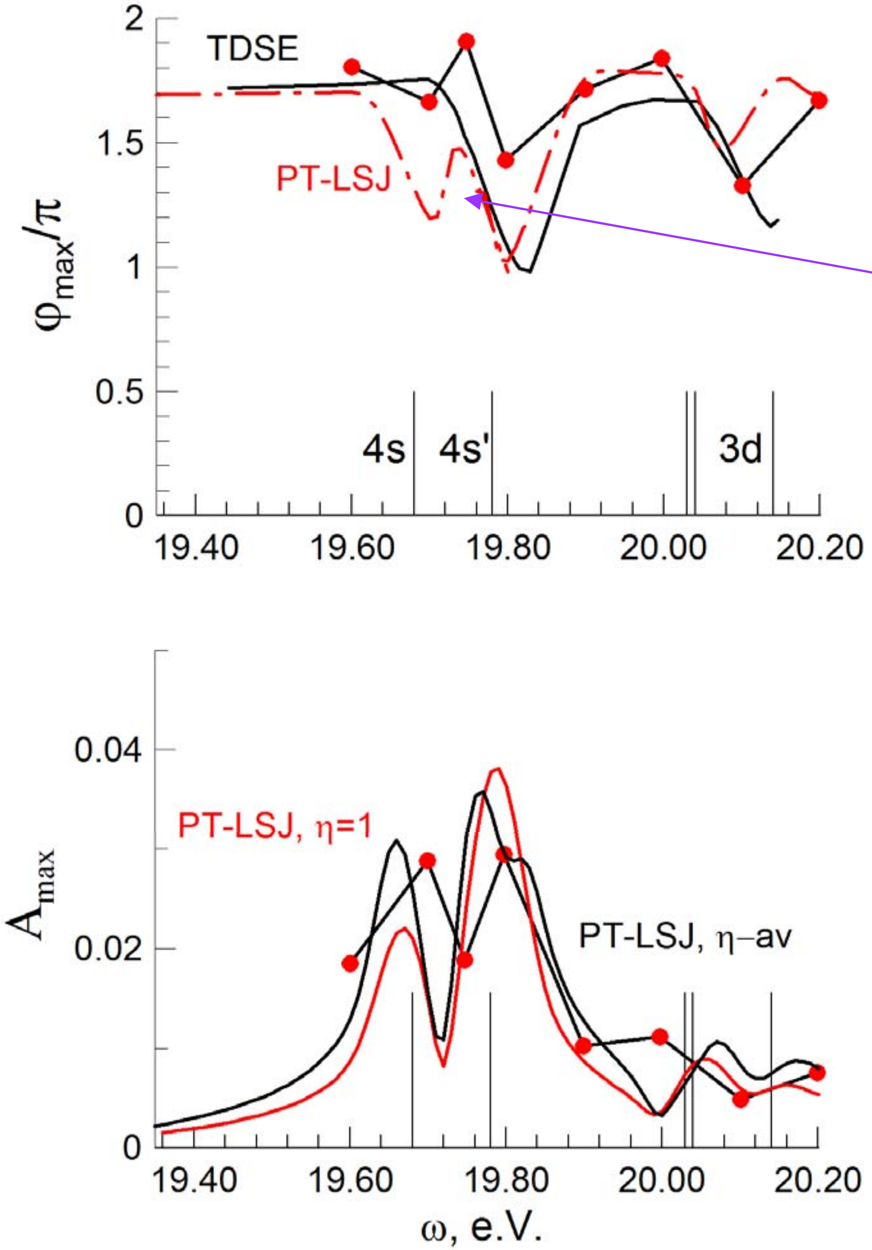
N=500
~100 fs





Weak resonance and large η lead to $\Gamma_\gamma > \Gamma_\beta$

Experimental data



Experimental data are provided by Giuseppe Sansone and collaborators

appearance of two S-matrix poles

Complete experiment?

Conventional single-photon ionization $2p^{5\,2}Pes\,^1P$, $2p^{5\,2}Ped\,^1P$
One complex ratio and one parameter β_2

Conventional two-photon ionization $2p^{5\,2}Pep\,^1S$, $2p^{5\,2}Pep\,^1D$, $2p^{5\,2}Pef\,^1D$
Two complex ratios and two parameters $\beta_{2,4}$

Bichromatic (ω+2ω) ionization $2p^{5\,2}Pep\,^1S$, $2p^{5\,2}Pep\,^1D$, $2p^{5\,2}Pef\,^1D$, $2p^{5\,2}Pes\,^1P$, $2p^{5\,2}Ped\,^1P$
Four complex ratios and six parameters $\beta_{2,4}, \left|\beta_{1,3}\right|, \phi_{1,3}^{(m)}$

Acknowledgments

A. N. Grum-Grzhimailo, E.I. Staroselskaya,

K. Bartschat, N. Douguet

K. C. Prince, E. Allaria, C. Callegari, R. Cucini, G. De Ninno,
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Y. Kumagai, T. Takanashi, **K. Ueda**, A. Fischer, M. Coreno,
F. Stienkemeier, Y. Ovcharenko, T. Mazza and M. Meyer

Concluding remarks

- Coherent control of photoelectron angular distribution at bichromatic ionization was analyzed for the case of single and double intermediate resonance (s)

- The role of temporal (longitudinal) coherency, and phase behavior of the asymmetry in vicinity of single and double resonances was investigated

- It is shown that with such type of experiment accompanied by conventional two-photon ionization one can perform a complete experiment, using only linearly polarized fields

Thank you for your attention!

Role of triplet states

$$\frac{dW}{d\Omega} = \frac{dW}{d\Omega}^{(I)} + \frac{dW}{d\Omega}^{(II)} + \frac{dW}{d\Omega}^{(III)}$$

$$\frac{dW}{d\Omega}^{(I)} = \frac{(-1)^{L_f}}{4\pi} \sum_{kll'} \hat{l}\hat{l}'(10,10\,|\,k0)(l0,l'0\,|\,k0) \begin{Bmatrix} l & l' & k \\ 1 & 1 & L_f \end{Bmatrix} e^{i(\Delta_l^{(1)} - \Delta_{l'}^{(1)})} D^{(0)}_{0\to L_f l,1} D^{(0)\,*}_{0\to L_f l',1} P_k(\cos\vartheta)\,,$$

$$\begin{aligned} \frac{dW}{d\Omega}^{(III)} &= \eta \frac{(-1)^{L_f}}{2\pi\sqrt{3}} \mathrm{Re}\Bigg[\sum_{ll'L'k} \hat{l}\hat{l}'(l0,l'0\,|\,k0)(10,L'0\,|\,k0)(10,10\,|\,L'0) \begin{Bmatrix} l & l' & k \\ L' & 1 & L_f \end{Bmatrix} \\ &\times e^{i(\Delta_l^{(1)} - \delta_{l'}^{(2)})} D^{(0)}_{0\to L_f l,1} T^{(1)} \left(\sum_n \left| \alpha_{10}^{\zeta_n} \right|^2 T^{(2)}_{E_n} D^{(0)\,*}_{n,1\to L_f l',L'} D^{(0)\,*}_{0\to n,1} \right) \Bigg] P_k(\cos\vartheta)\,, \end{aligned}$$

$$\begin{aligned} \frac{dW}{d\Omega}^{(II)} &= |\eta|^2 \frac{(-1)^{L_f}}{4\pi} \sum_{ll'JJ'k} \hat{l}\hat{l}'\hat{J}\hat{J}'(l0,l'0\,|\,k0)(10,10\,|\,J0)(10,10\,|\,J'0)(J0,J'0\,|\,k0) \\ &\times\ e^{i(\Delta_l^{(2)} - \Delta_{l'}^{(2)})} \sum_{SLL'} (-1)^{S+L+L'} \hat{L}\hat{L}' \begin{Bmatrix} l & l' & k \\ L' & L & L_f \end{Bmatrix} \begin{Bmatrix} L' & L & k \\ J & J' & S \end{Bmatrix} \\ &\times \left(\sum_{n,L_n} \alpha^{\zeta_n}_{L_n S} \alpha^{\zeta_n\,*}_{10} T^{(2)}_{E_n} \begin{Bmatrix} S & L & J \\ 1 & 1 & L_n \end{Bmatrix} D^{(S)}_{n,L_n\to L_f l,L} D^{(0)}_{0\to n,1} \right) \\ &\times \left(\sum_{n',L_{n'}} \alpha^{\zeta_{n'}\,*}_{L_{n'} S} \alpha^{\zeta_{n'}}_{10} T^{(2)}_{E_{n'}} \begin{Bmatrix} S & L' & J' \\ 1 & 1 & L_{n'} \end{Bmatrix} D^{(S)\,*}_{n',L_{n'}\to L_f l',L'} D^{(0)\,*}_{0\to n',1} \right) P_k(\cos\vartheta)\,, \end{aligned}$$

Interference is possible only for S=S'