Dynamics of angular patterns in two-photon sequential double ionization of neon

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QUAMP, Swansea 08-12 Sept 2013

Outline

1. Two-photon ionization of neon from valence np6-shell:

> Brief description of the process

> Some representative experiments

2. Set-up of the problem and discussion:

> Two-electron angular patterns under 44 eV & 50 eV photons

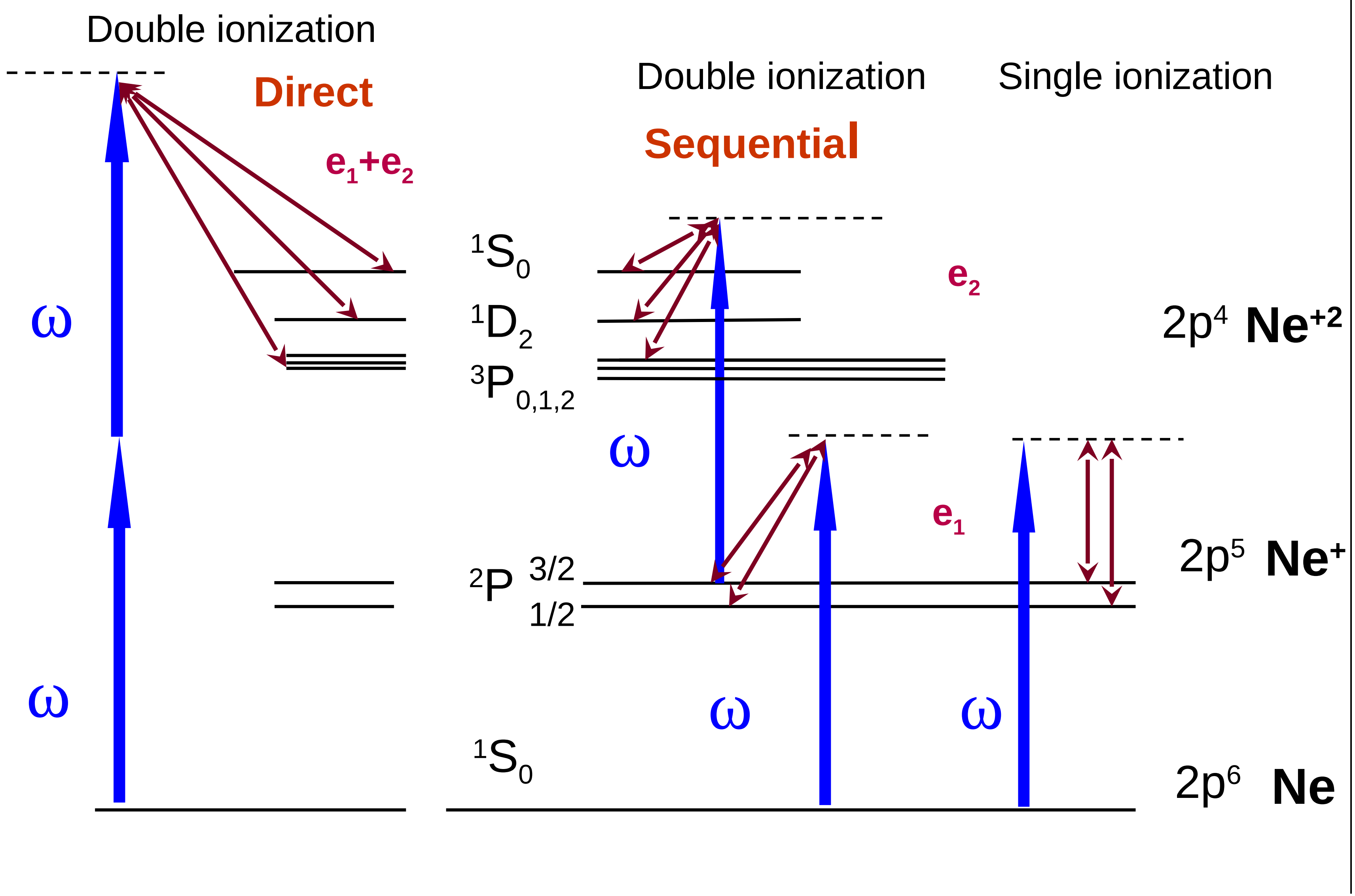
3. Dynamics of the angular patterns

>Two-electron photoelectron angular distributions (**PADs)**

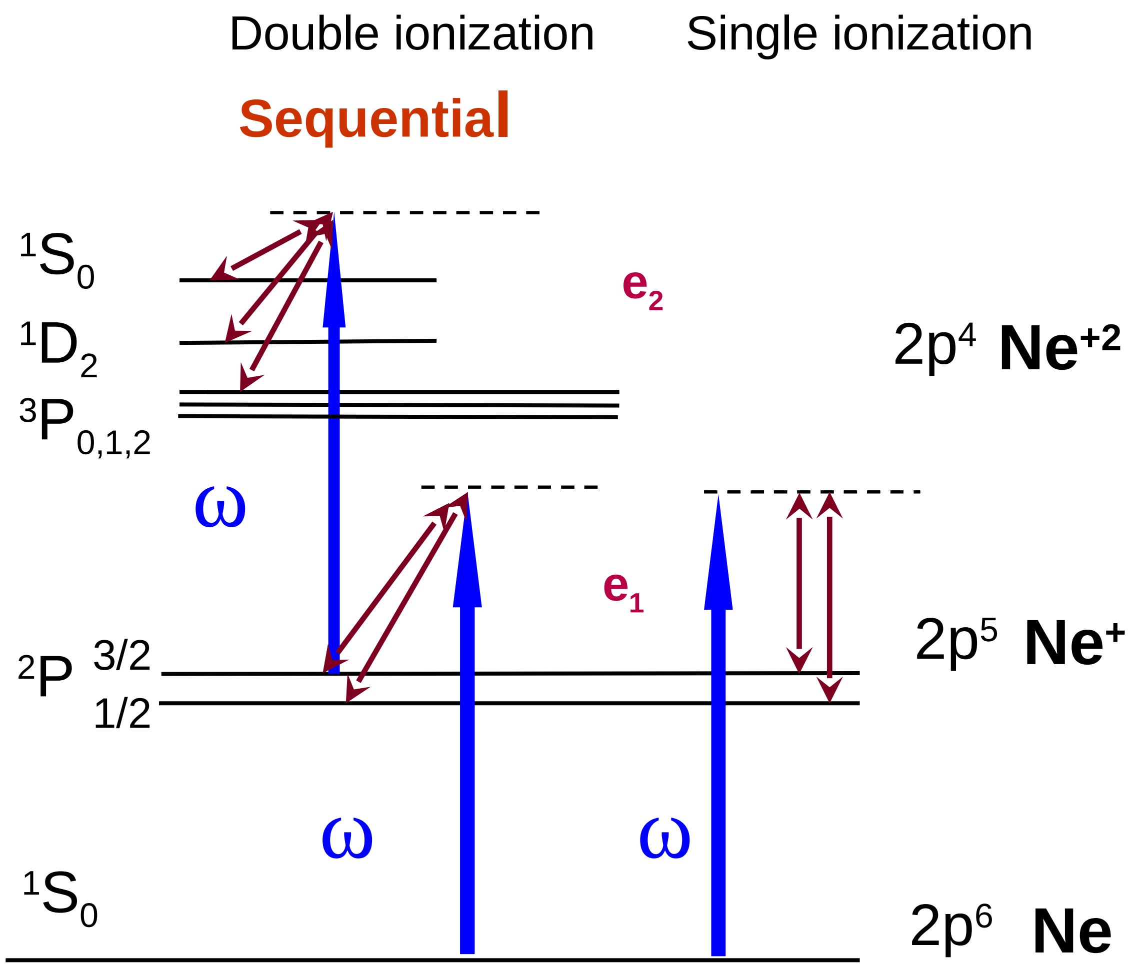
> Ionic dynamic alignment

Two-photon ionization of neon

Two-photon ionization of neon



photon energy > 41 eV : sequential path

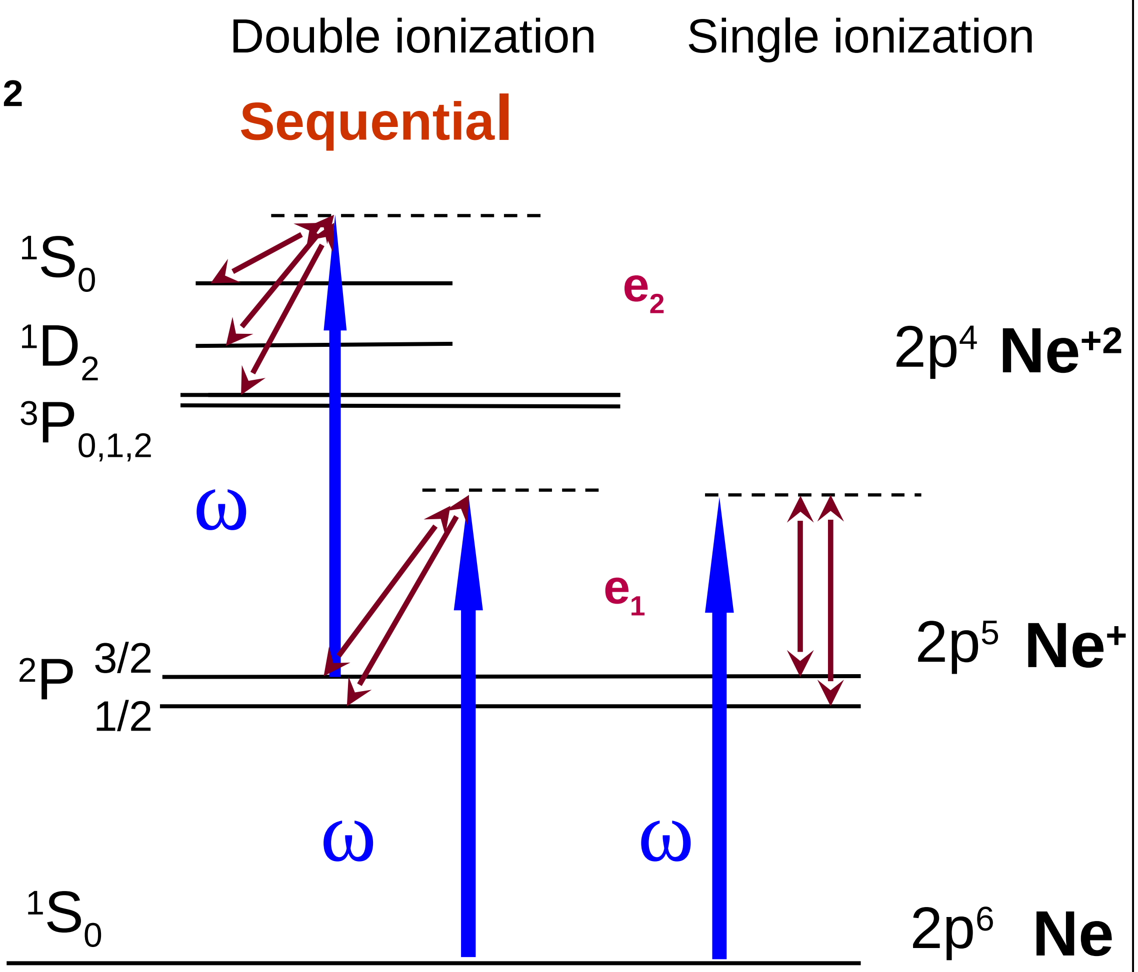


Sequential double ionization path dominates

$$E^0 + 2\omega = E^{2+} + e_1 + e_2$$

$$E^{1+} + \omega = E^{2+} + e_2$$

$$E^0 + \omega = E^{1+} + e_1$$



Works of interest for this talk

1. ***M. Braune, A. Reinkoster, J. Viefhaus, B. Lohmann and U. Becker, 25th ICOMP, Freiburg, 2007*** ***(experiment)***

2. **JPB 42, 141002 (2009), M. Kurka *et al*** ***(Experiment)***

'Two-photon double ionization of Ne by FEL radiation: a kinematically complete experiment'

3. **JPB, 41, 165601 (12pp) (2008), S. Fritzsche, A.N. Grum-Grzhimailo, E.V. Gryzlova, N.M. Kabachnik** **(Theory)**

'Angular distributions and angular correlations in sequential two-photon double ionization of atoms'

4. **JPB, 42, 134016, (2009) A. S. Kheifets** **(Theory)**

' *Photoelectron angular correlation pattern in sequential two-photon double ionization of neon*.'

FEL radiation and measurements

Experiment (1) : M. Braune et al, 2007

47.5 eV, Neon and Argon, non-coincident measurements

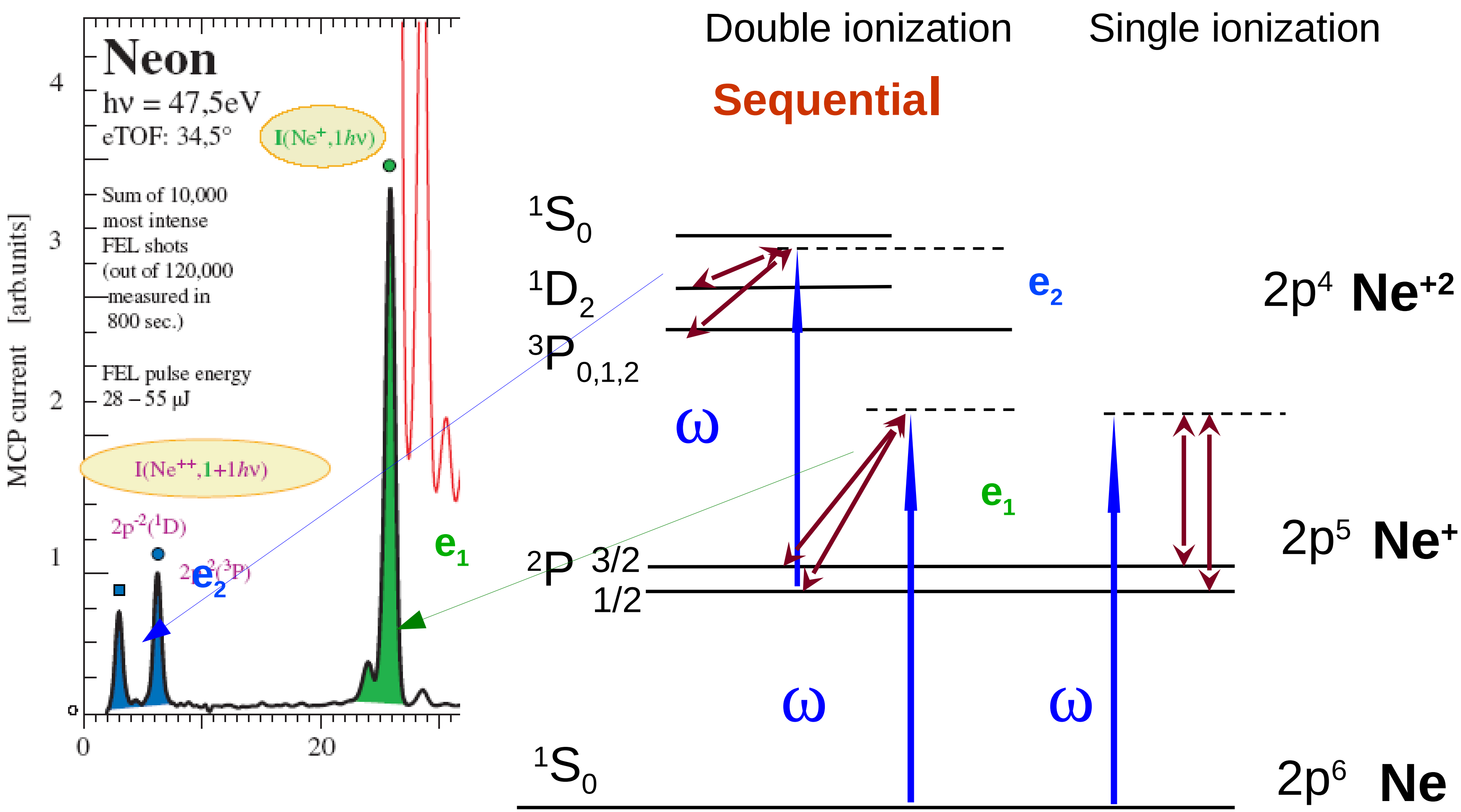
> photoelectron energy spectrum of the electrons
> asymmetry parameters for 2nd-step electron

Experiment (2) : Kurka *et al*, 2009

44 eV, ~25 fs, ~ 5 x 10^{13} W/cm^2

> photoelectron energy spectrum
> asymmetry parameters for non-coincident measurements
> angular patterns for coincident measurements
> Comparison with theory.

Photoelectron energy spectrum

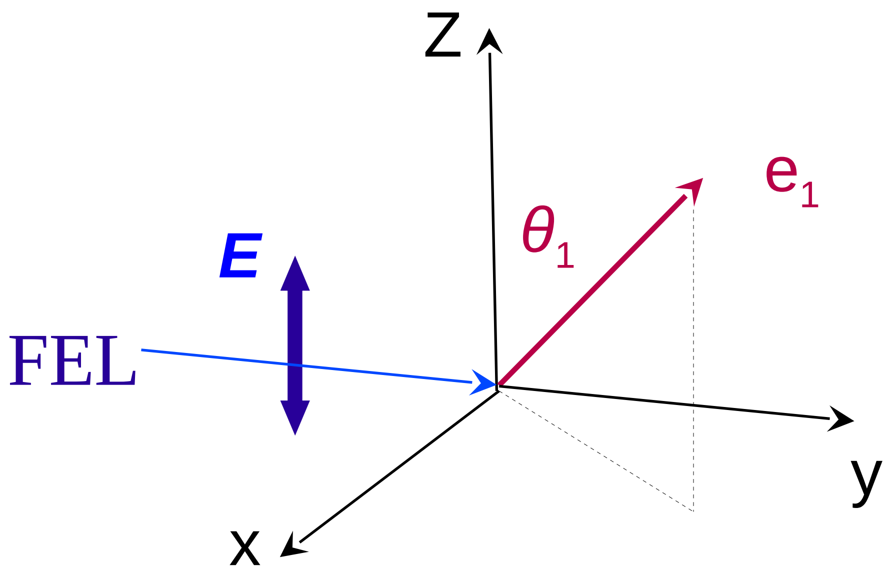


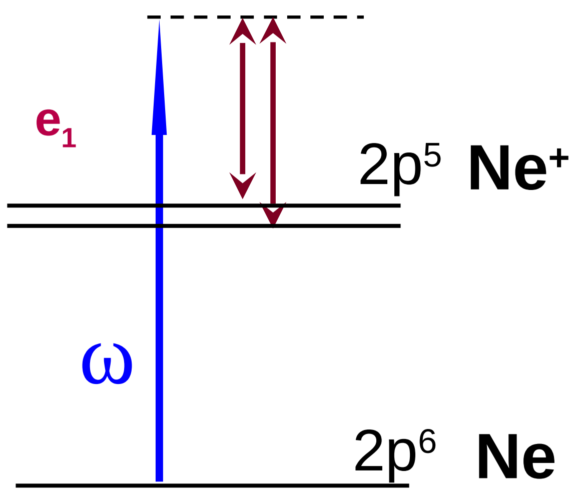
(1) M. Braune, et al 2007

Single Ionization: angular distribution of electron

Linearly polarized XUV radiation, Z ∥ ***E***

Single ionization



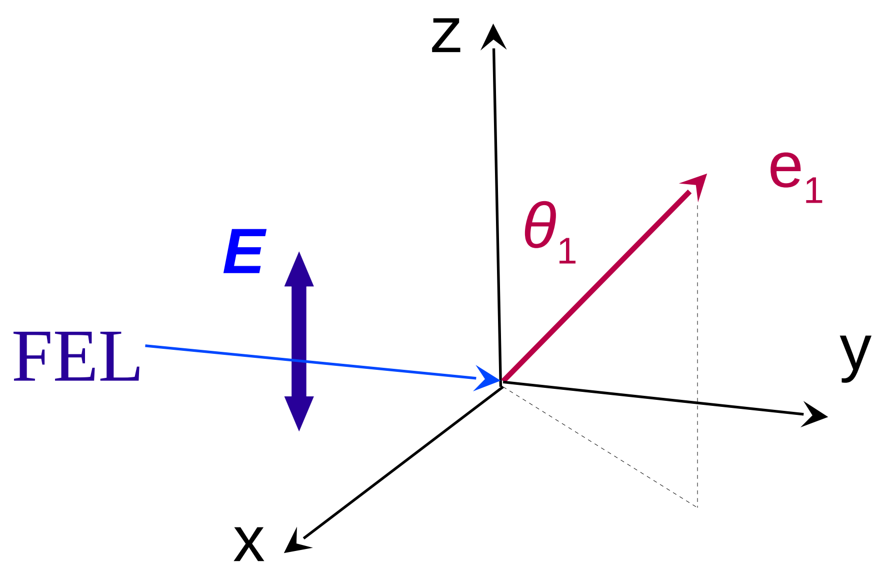


$W_1 \sim 1 + \beta_2 P_2(\cos\theta_1)$ **Angular distribution of photoelectron e_1**

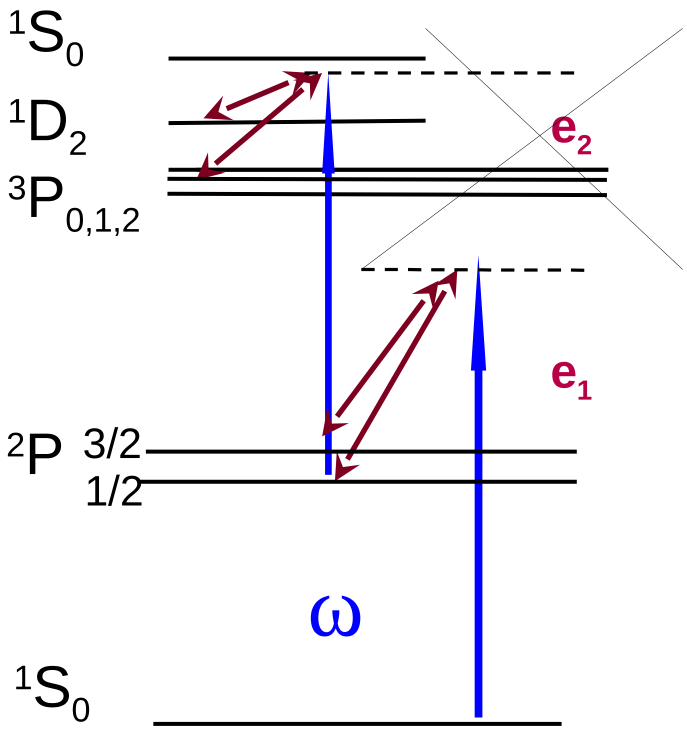
β_2 Asymmetry parameter (deviation from spherical shape)

Double ionization: angular distribution of the 1st electron

Linearly polarized XUV radiation, Z ‖ ***E***



Double ionization

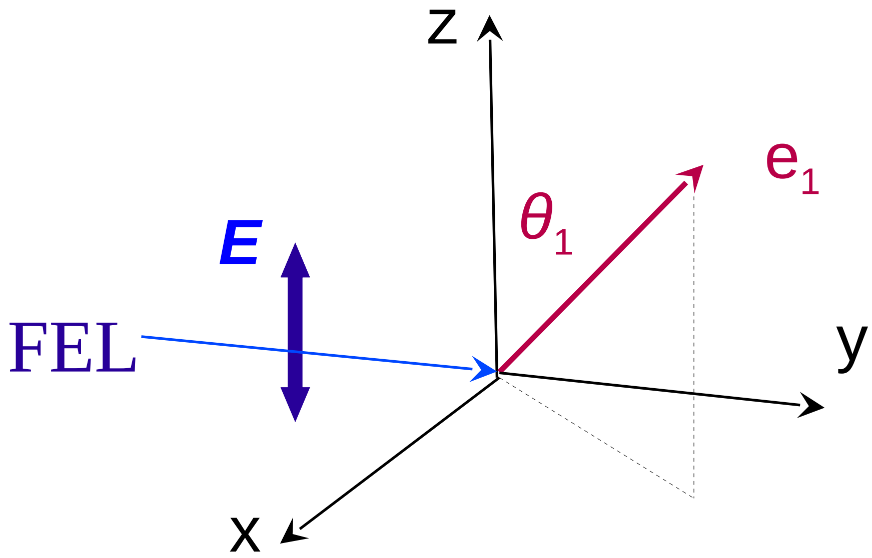


e_2 remains unobserved: (non-coincident measurement)

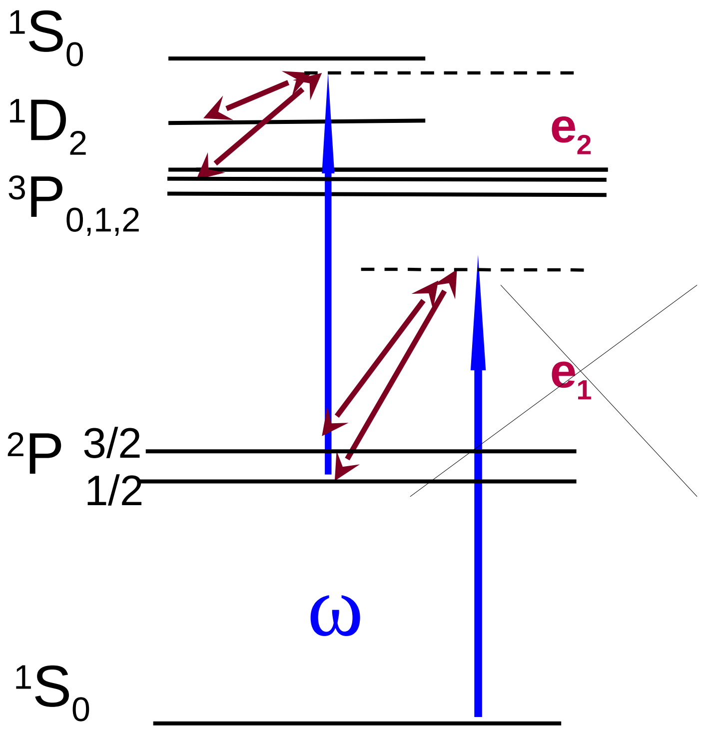
$W_1 \sim 1 + \beta_2 P_2(\cos\theta_1) + \beta_4 P_4(\cos\theta_1)$ **Angular distribution of photoelectron e_1**

Double ionization: angular distribution of the 2nd electron

Linearly polarized XUV radiation, Z ‖ ***E***



Double ionization

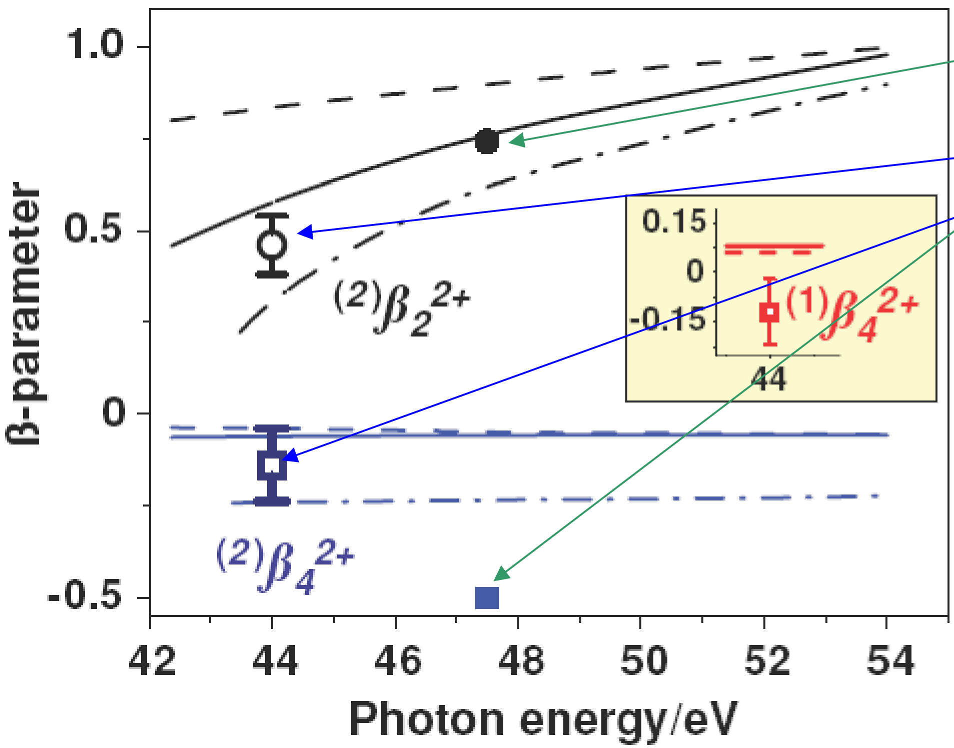


e$_1$ remains unobserved: (non-coincident measurement)

$W_2 \sim 1 + \beta'_2 P_2(\cos\theta_2) + \beta'_4 P_4(\cos\theta_2)$ **Angular distribution of photoelectron e$_2$**

Asymmetry parameters of the 2nd step electron

Final state: Ne^{+2} 3P (1st- electron not observed)



Braune et al, 2007, 47.5 eV

Kurka et al, 2009, 44 eV

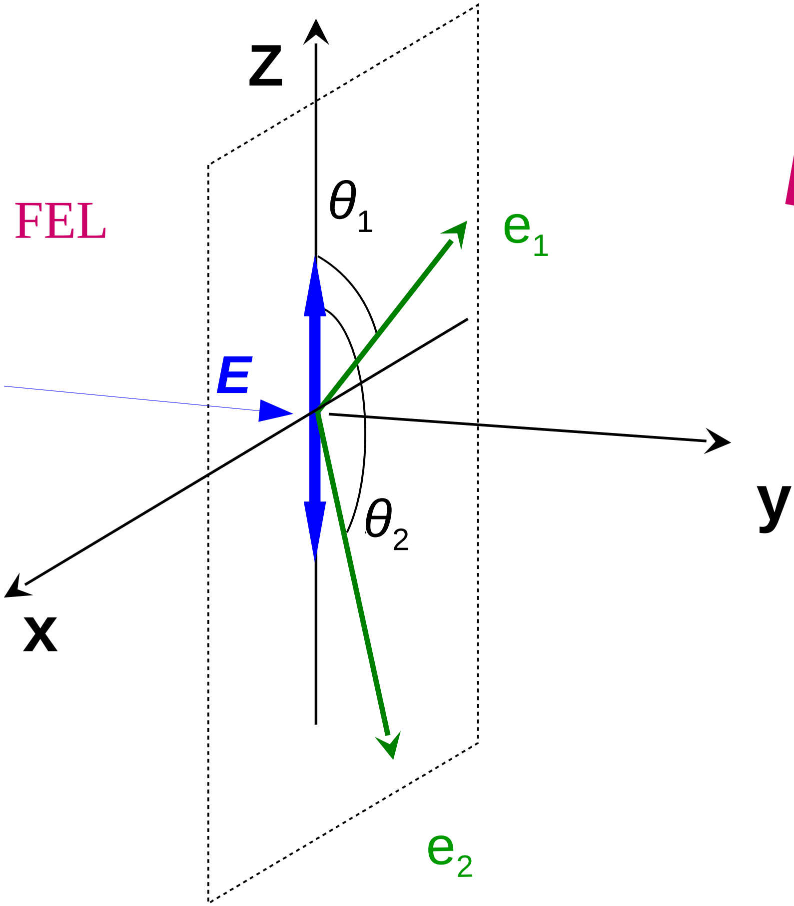
(Fritzsche et al, 2008; Kurka et al, 2009)

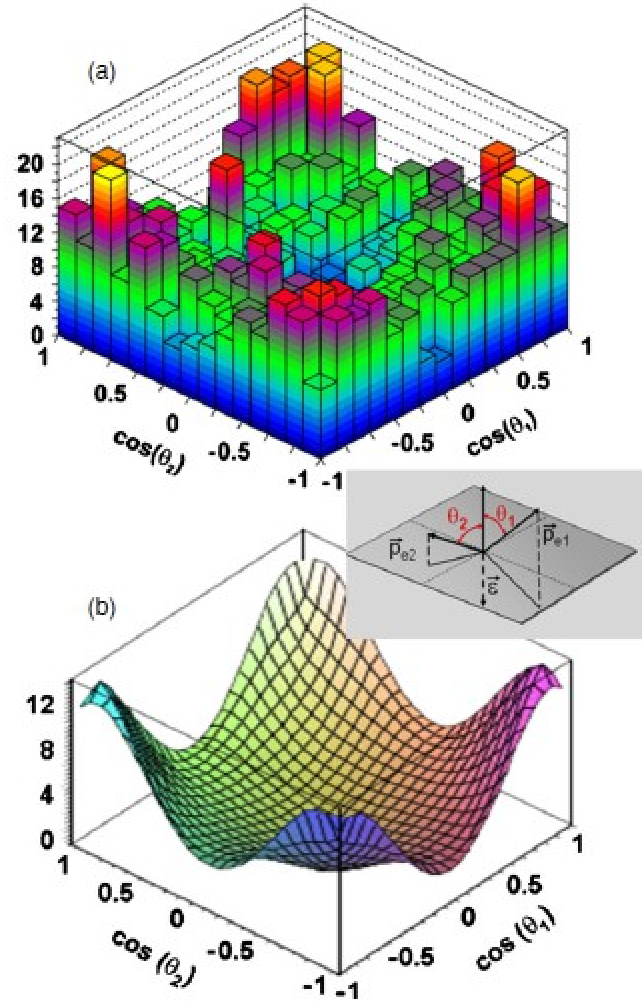
Kheifets, 2007

$\beta_2^{(2)}$ $\beta_4^{(2)}$

Two-electron angular distributions on the XZ-plane

FLASH 44 eV, ~25 fs, ~5·10^{13} W/cm^2





$$W(\theta_1, \phi_1=0; \theta_2, \phi_2=0)$$

Kurka et al, (2009)

Figure 3. Experimental (a) and theoretical (b) probability density distribution of two emitted electrons as a function of (cos(θ_1), cos(θ_2)), where θ is the emission angle with respect to the FLASH polarization direction. Inset: sketch of the experimental geometry.

Two-electron angular distributions

The problem is that the two theoretical formulations give different predictions for the 2-electron angular patterns.

?

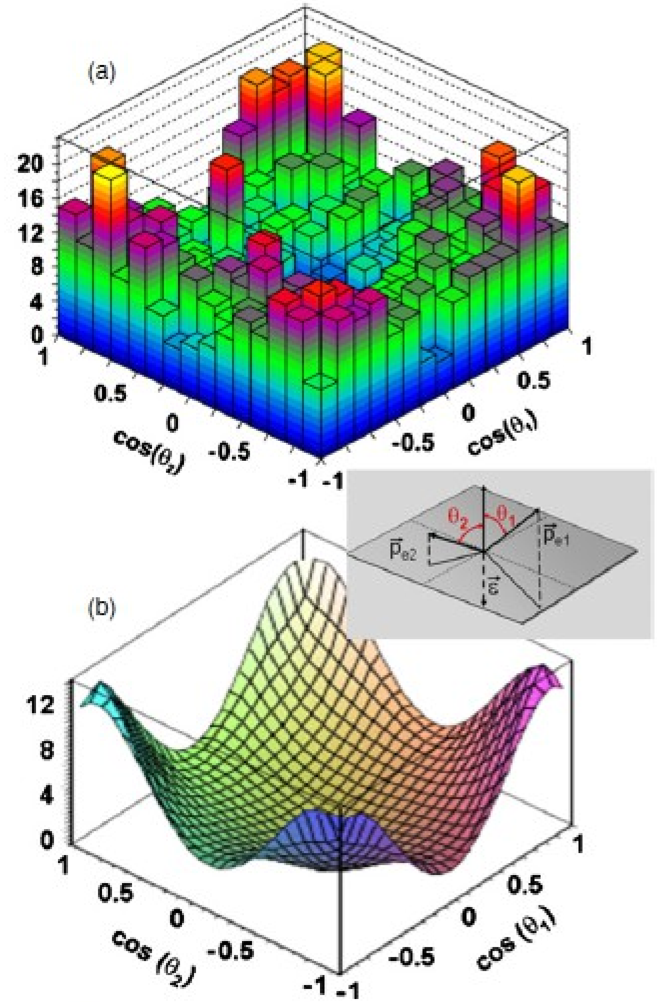
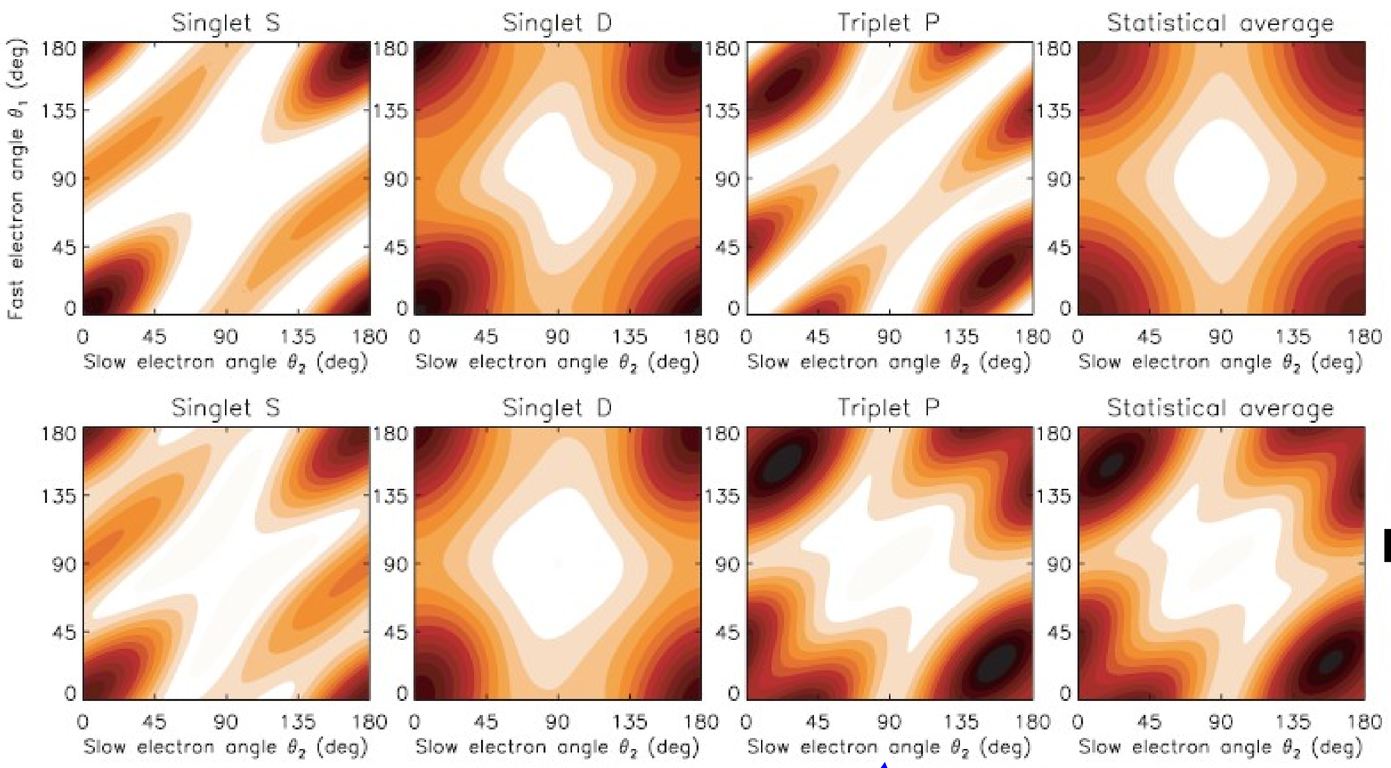


Figure 3. Experimental (a) and theoretical (b) probability density distribution of two emitted electrons as a function of ($\cos(\theta_1)$, $\cos(\theta_2)$), where θ is the emission angle with respect to the FLASH polarization direction. Inset: sketch of the experimental geometry.

Two-electron angular distributions: theory at 50 eV



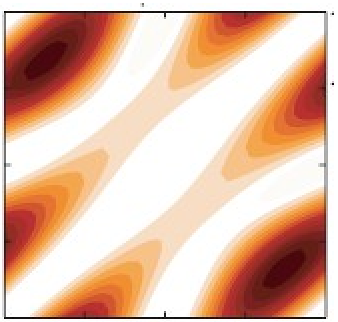
A. kheifets 2009

Fritzsche et al, 2008

Maulbetsch and Briggs J.Phys. B, 28, 551 (1995)

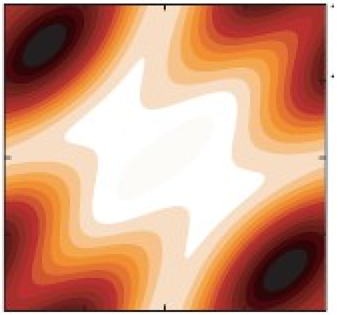
³P final state: emission of electrons along the polarization axis is forbidden

coherent versus incoherent excitation of the fine structure intermediate ionic levels



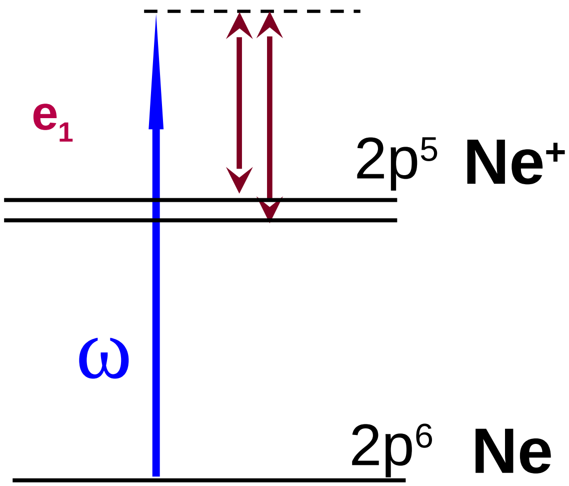
coherent
LSM_sM_L

Incoherent
JM_J



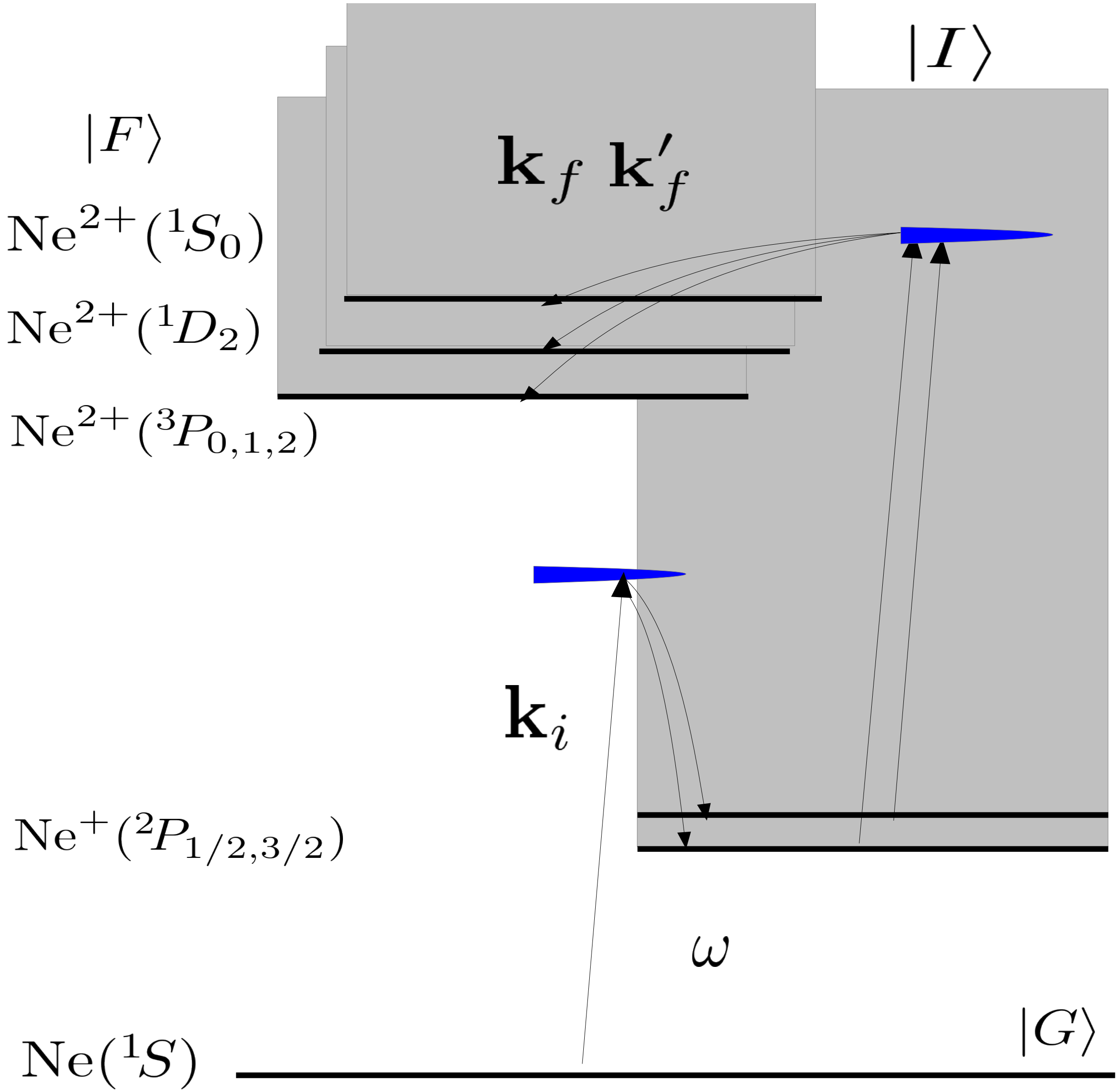
A. Kheifets 2009

Fritzsche et al, 2008

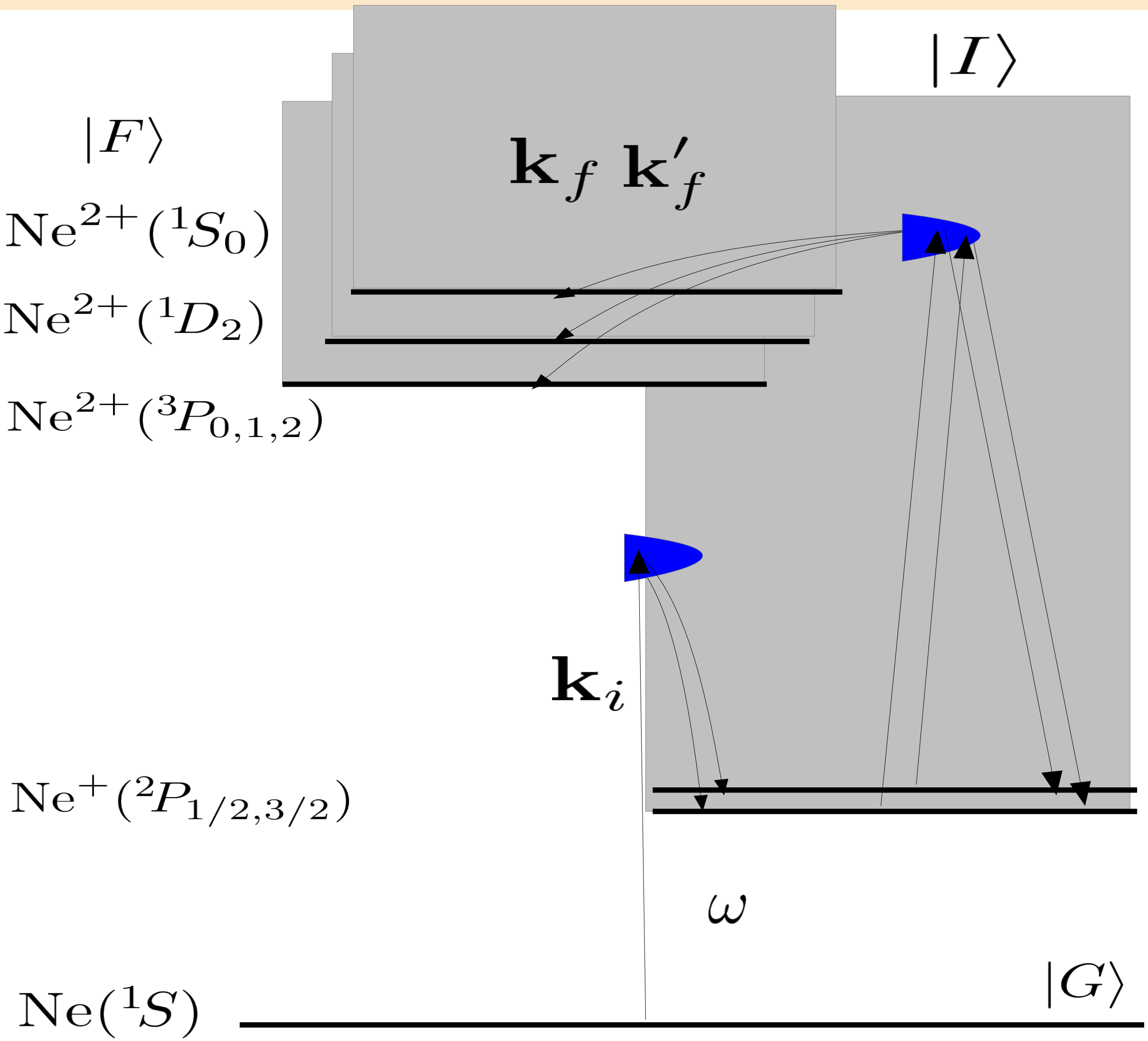


E_{SO} ~ 0.1 eV ----> .42.7 fs precession period

Long pulse / incoherent excitation



Short pulse / coherent excitation



Raman transitions

Short pulse / coherent excitation

Much shorter pulses excite the doublet coherently!

Also allow for Raman transitions (strong-field effects)

A time-dependent approach of the problem is required for a consistent theory!

Details of the theory will be skipped at the moment

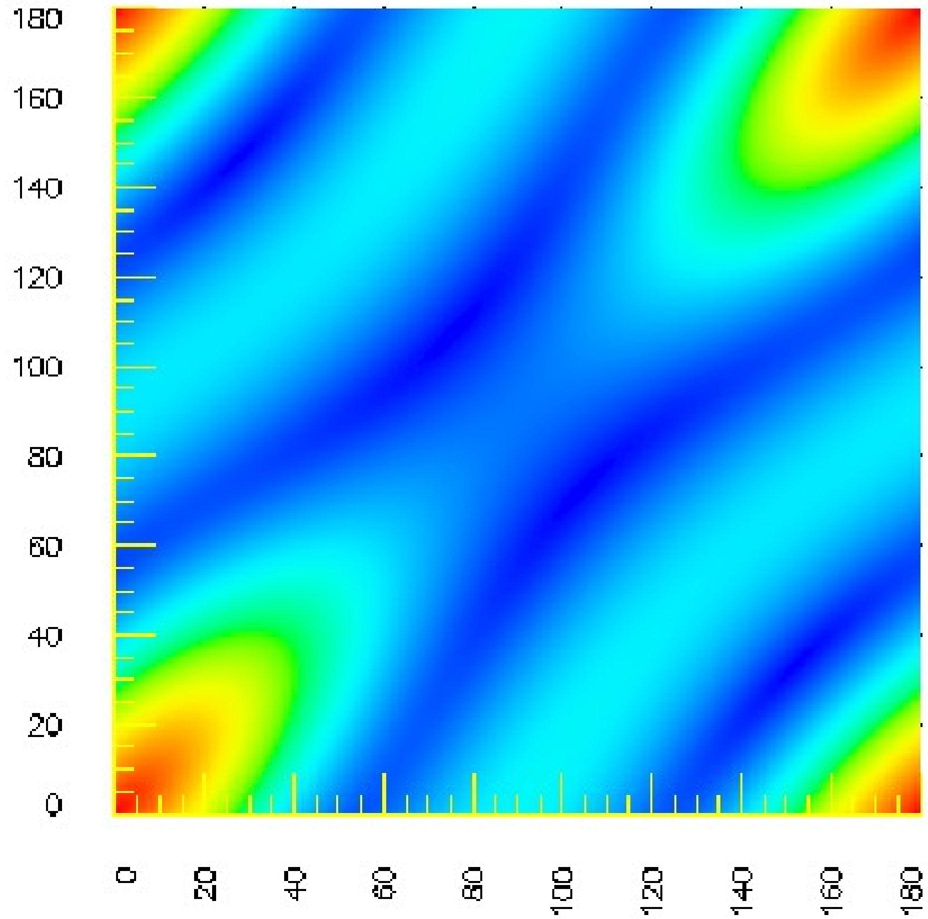
Results

1. The effect of the pulse duration on the angular patterns

Two-electron angular pattern

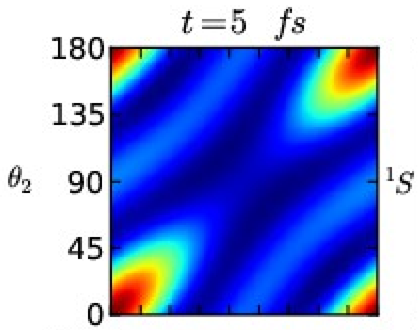
$$W(\theta_1, \phi_1=0; \theta_2, \phi_2=0)$$

Angle of 1st electron

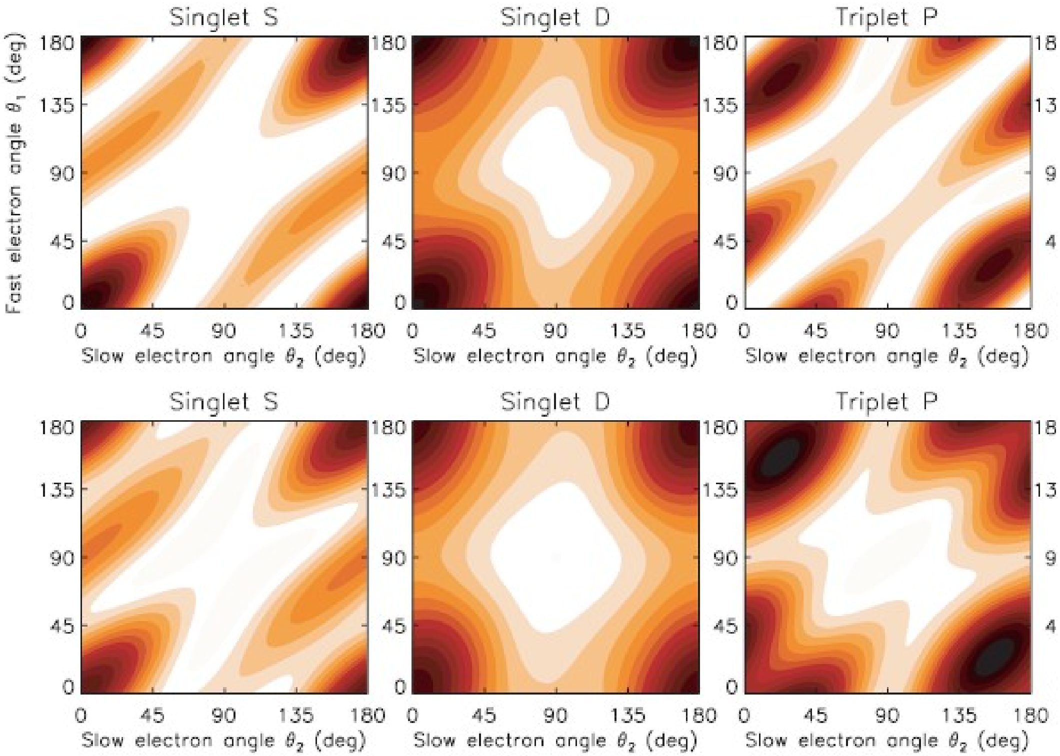


Angle of 2nd-step electron

Two-electron angular patterns

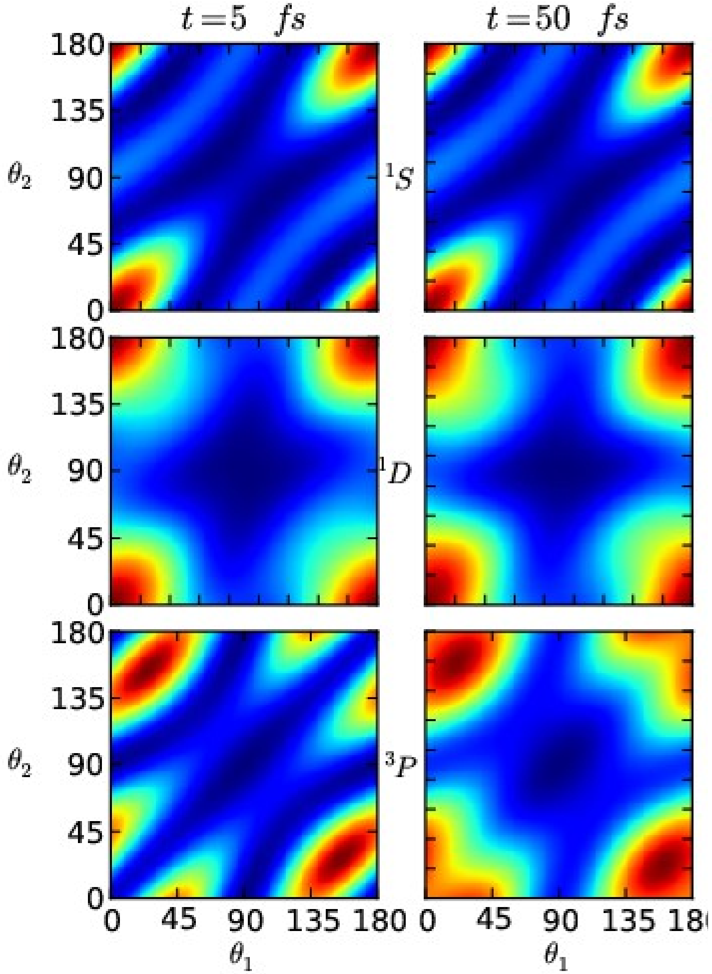


A. kheifets 2009 / coherent summation



Fritzsche et al, 2008 / incoherent summation

Two-electron angular patterns



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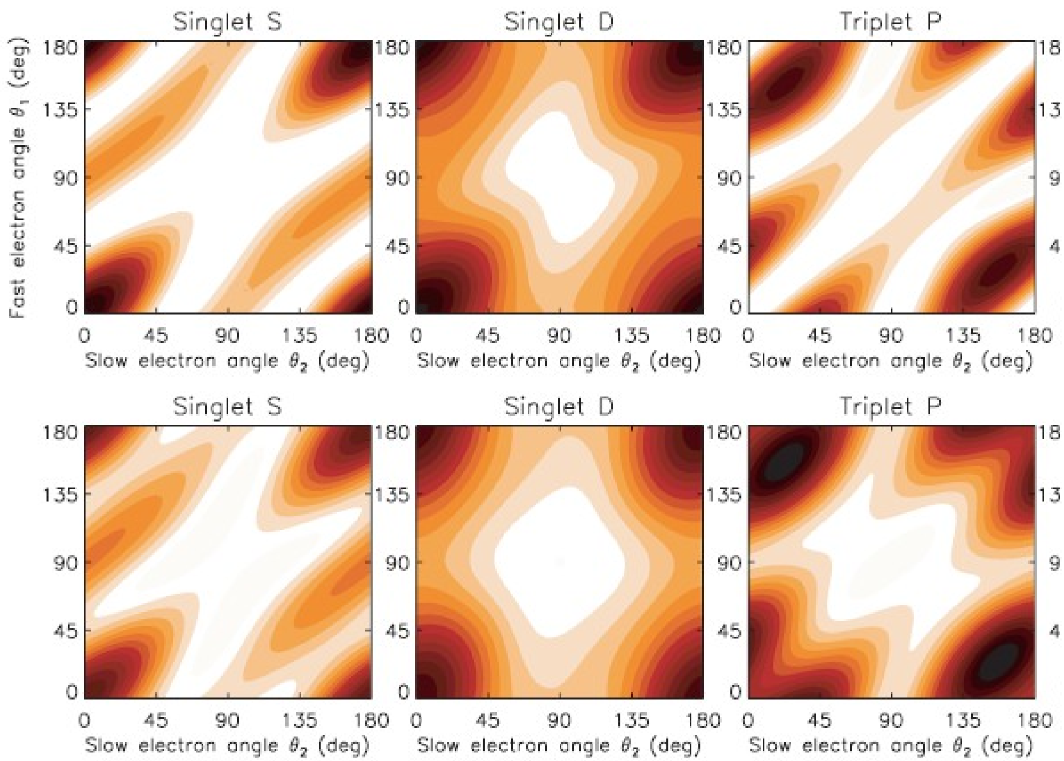


Fritzsche et al, 2008 / incoherent summation

Dynamic alignment

2. Strong field effects on the ionic dynamic alignment

Dynamic alignment of the singly charged neon

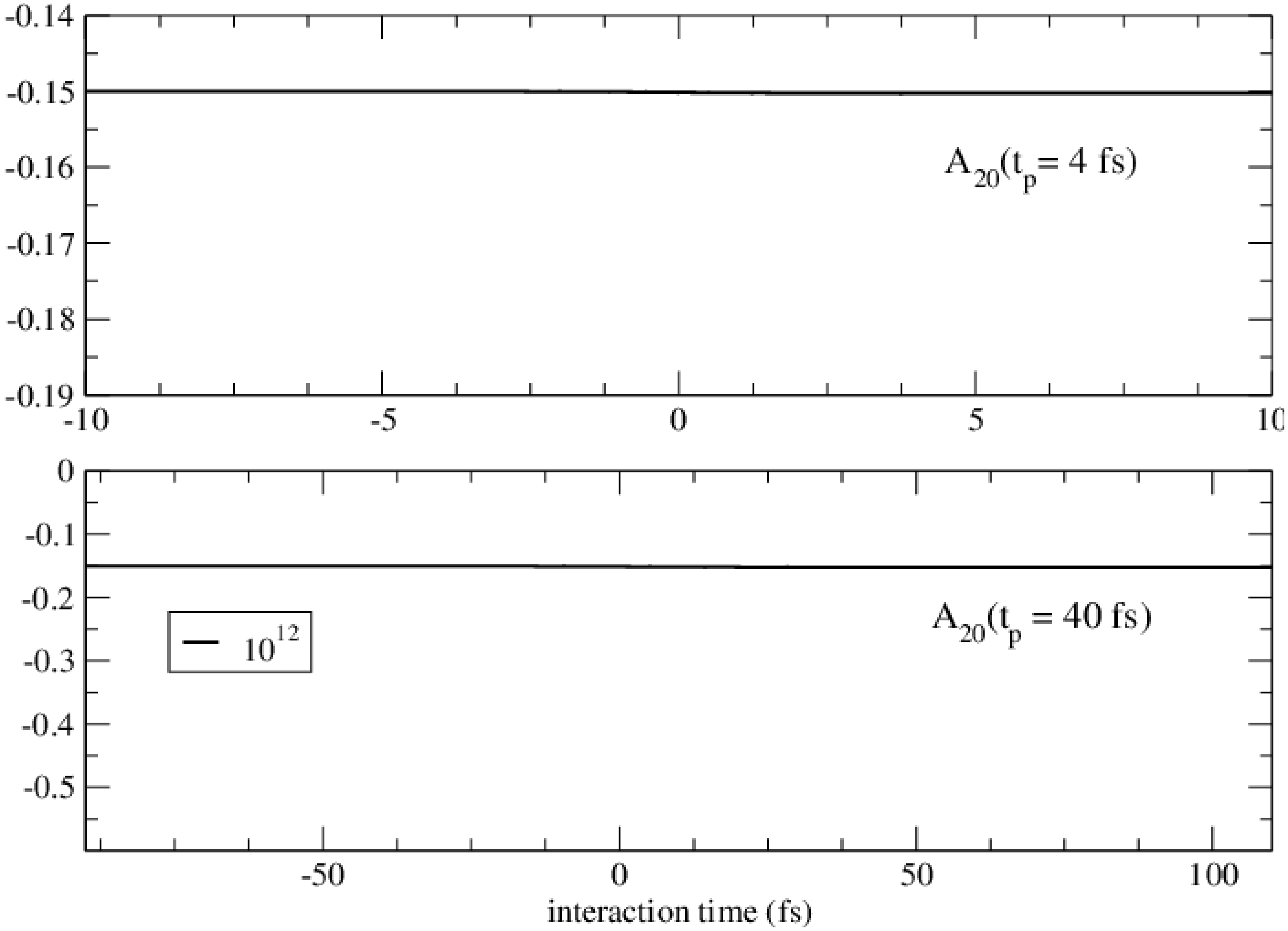
Alignment

The magnetic substates of the intermediate Ne+ ionic states are not equally populated: The populations of M_J = 1/2 and M_J = 3/2 of the J = 3/2 state of the spin-orbit double differ.

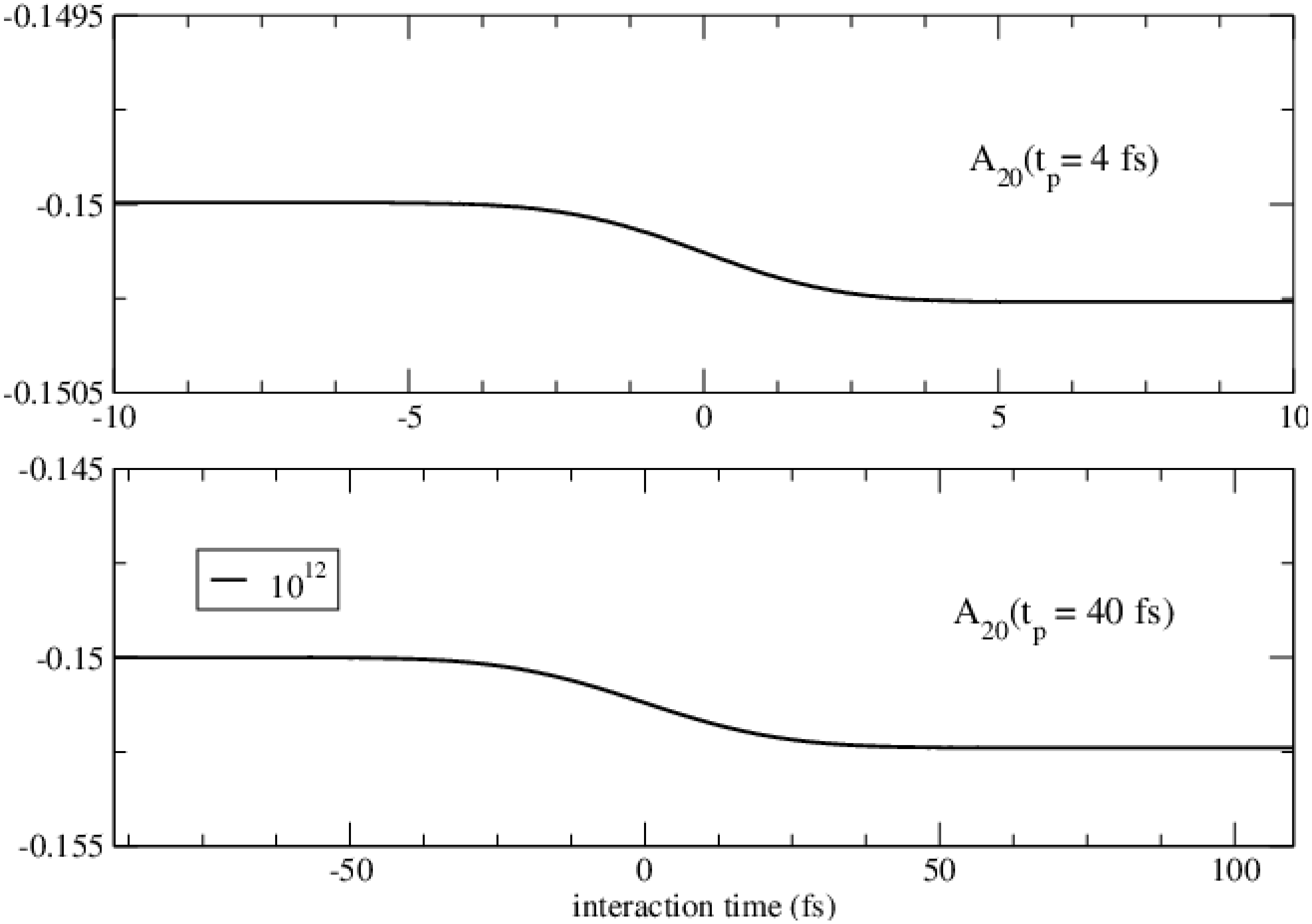
$$A_{20} = [\, P(3/2) - P(1/2) \,] \;/\; [\, P(3/2) + P(1/2) \,]$$

(For Neon standard photoionization theory gives A_{20} = - 0.15)

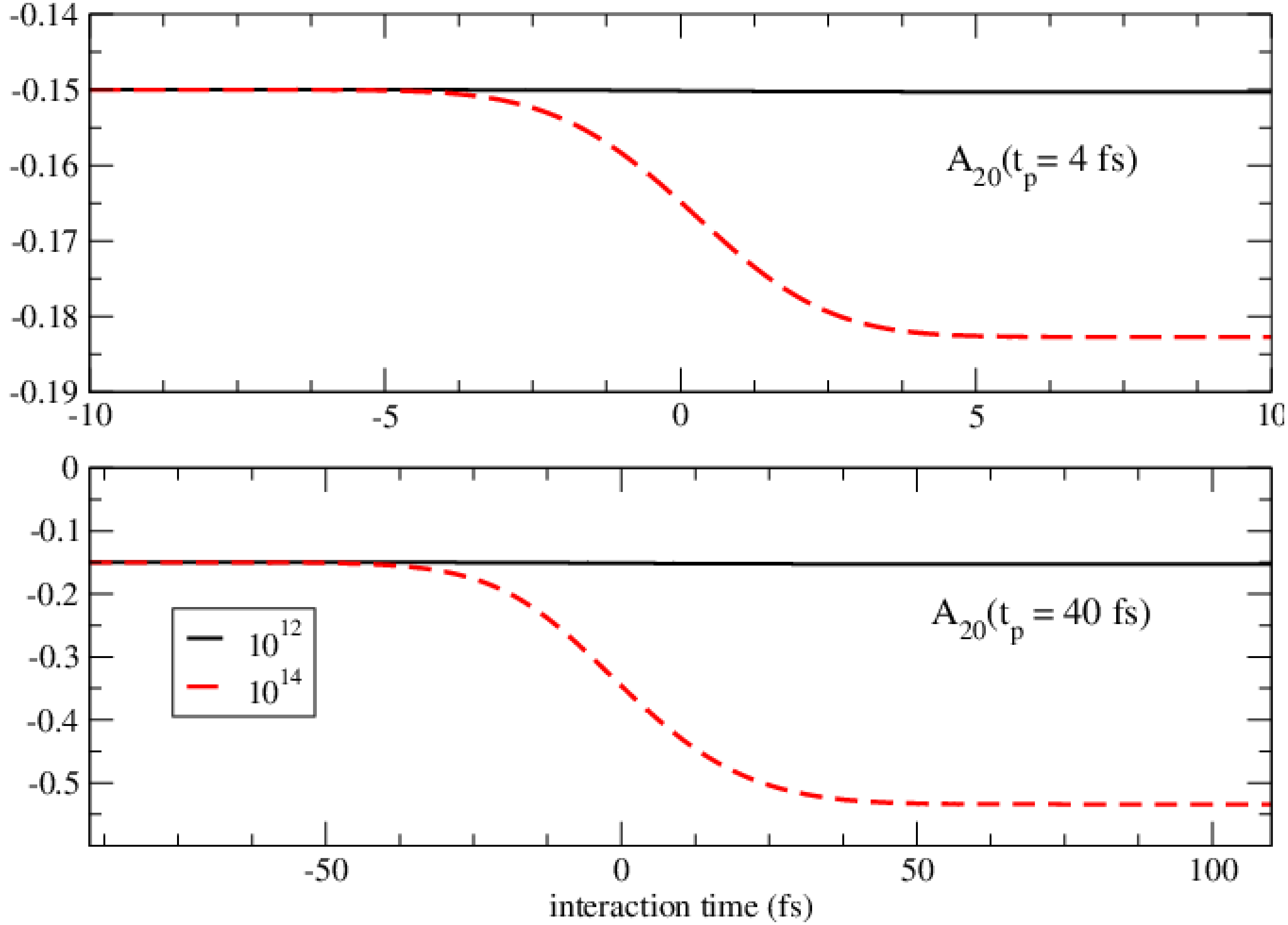
Dynamic alignment of the singly charged neon



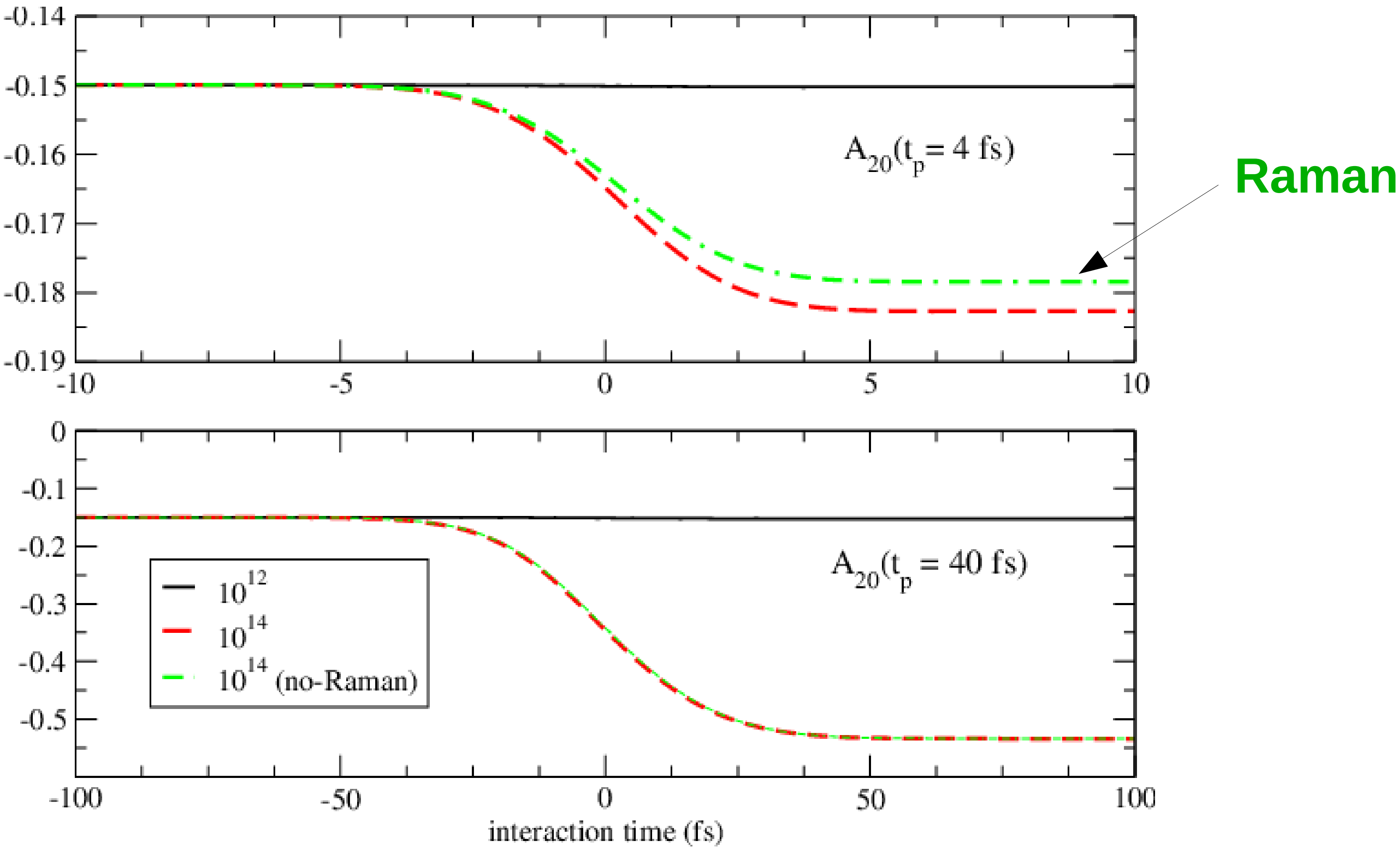
Dynamic alignment of the singly charged neon



Dynamic alignment of the singly charged neon



Dynamic alignment of the singly charged neon



Summary & Conclusions

✔

1. **A time-dependent approach followed for the description of the two-photon sequential double ionization of neon. (PRL 111, 093001, (2013)**

2. **Pulse duration and/or strong field effects important for the interpretation of reported experimental results**

3. **Bound state dynamics (coherent or incoherent time evolution of the residual ion, spin-orbit precession) decisively determine angular correlations**