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

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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

Double ionization

Direct

Sequential

\( e_1 + e_2 \)

\( \omega \)

\( 2p^4 \) Ne\(^{+2} \)

\( 2p^5 \) Ne\(^+ \)

\( 2p^6 \) Ne
photon energy > 41 eV: sequential path

Sequential

$^1S_0$

$^1D_2$

$^3P_{0,1,2}$

$^2P_{3/2,1/2}$

$^1S_0$

Double ionization

Single ionization

$2p^4 \text{ Ne}^{+2}$

$2p^5 \text{ Ne}^+$

$2p^6 \text{ Ne}$
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


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'


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

Neon
hv = 47.5 eV
eTOF: 34.5°

Sum of 10,000 most intense FEL shots (out of 120,000 measured in 800 sec.)
FEL pulse energy 28 – 55 μJ

I(Ne⁺,1hv)
I(Ne⁺⁺,1+1hv)

2p²(1D)
2p³(3p)

2p⁴ Ne⁺⁺
2p⁵ Ne⁺
2p⁶ Ne

(1) M. Braune, et al 2007
Single Ionization: angular distribution of electron

Linearly polarized XUV radiation, $Z \parallel E$

Single ionization

$W_1 \sim 1 + \beta_2 P_2(\cos \theta_1)$

Angular distribution of photoelectron $e_1$

$\beta_2$ Asymmetry parameter (deviation from spherical shape)
Double ionization: angular distribution of the 1st electron

Linearly polarized XUV radiation, $Z \parallel E$

$e_2$ 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, $Z \parallel E$

$e_1$ 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 2\textsuperscript{nd} step electron

Final state: Ne\textsuperscript{+2} 3P  \hspace{1em} (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

\[ \beta_2^{(2)} \quad \beta_4^{(2)} \]
Two-electron angular distributions on the XZ-plane

FLASH

$44 \text{ eV, } \sim 25 \text{ fs, } \sim 5 \cdot 10^{13} \text{ W/cm}^2$

FEL

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

Kurka et al, (2009)

Figure 3. Experimental (a) and theoretical (b) probability density distribution of two emitted electrons as a function of $(\cos(\theta_1), \cos(\theta_2))$, where $\theta$ is the emission angle with respect to the FLASH polarization direction. Inset: sketch of the experimental geometry.
The problem is that the two theoretical formulations give different predictions for the 2-electron angular patterns.
Two-electron angular distributions: theory at 50 eV

- Fritzschke et al., 2008
- A. Kheifets, 2009

$^3P$ final state: emission of electrons along the polarization axis is forbidden.
coherent versus incoherent excitation of the fine structure intermediate ionic levels

coherent $\text{LSM}_s M_L$  
Incoherent $\text{JM}_J$

$\text{Ne}^{+} 2p^5 \rightarrow 2p^6 \text{Ne} \quad E_{so} \sim 0.1 \text{ eV} \rightarrow .42.7 \text{ fs precession period}$

A. Kheifets 2009

Fritzsche et al, 2008
Long pulse / incoherent excitation

\[ |F\rangle \]
\[ \text{Ne}^{2+}(^1S_0) \]
\[ \text{Ne}^{2+}(^1D_2) \]
\[ \text{Ne}^{2+}(^3P_{0,1,2}) \]

\[ |k_f\rangle \quad |k'_f\rangle \]

\[ \text{Ne}^+(^2P_{1/2,3/2}) \]

\[ \text{Ne}(^1S) \]

\[ |I\rangle \]

\[ |G\rangle \]

\[ \omega \]
Short pulse / coherent excitation

\[ |F\rangle \]
\[ \text{Ne}^2+ (^{1S}_0) \]
\[ \text{Ne}^2+ (^{1D}_2) \]
\[ \text{Ne}^2+ (^{3P}_{0,1,2}) \]
\[ k_f \quad k'_f \]
\[ |I\rangle \]

Raman transitions

\[ \text{Ne}^+ (^{2P}_{1/2,3/2}) \]

\[ \text{Ne} (^{1S}) \]

\[ \omega \]

\[ |G\rangle \]
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
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

A. Kheifets 2009 / coherent summation

Fritzsche et al, 2008 / incoherent summation
2. Strong field effects on the ionic dynamic alignment
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.

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

(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

\[ A_{20}(t_p = 4 \text{ fs}) \]

\[ A_{20}(t_p = 40 \text{ fs}) \]

Raman
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