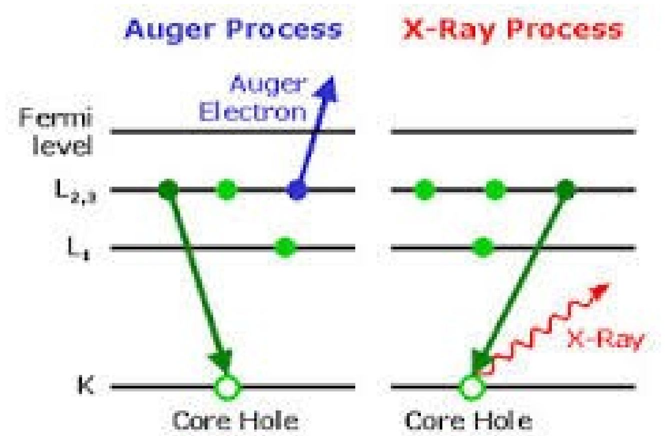
I. Role of shake-up transitions on the Auger cascades of light and medium elements



From: wiki.utep.edu

Electron emission from (inner-shell) excited states:

A^* →

$A^{+(*)}$ + e^-_{Auger}	dominant process
→ A^{++} + e^-_{Auger} + …	Auger cascades
$A^{++(*)}$ + $e^-_{Auger,1}$ + $e^-_{Auger,2}$	double Auger decay
$A^{++(*)}$ + e^-_{Auger} + $h\omega$	radiative Auger decay

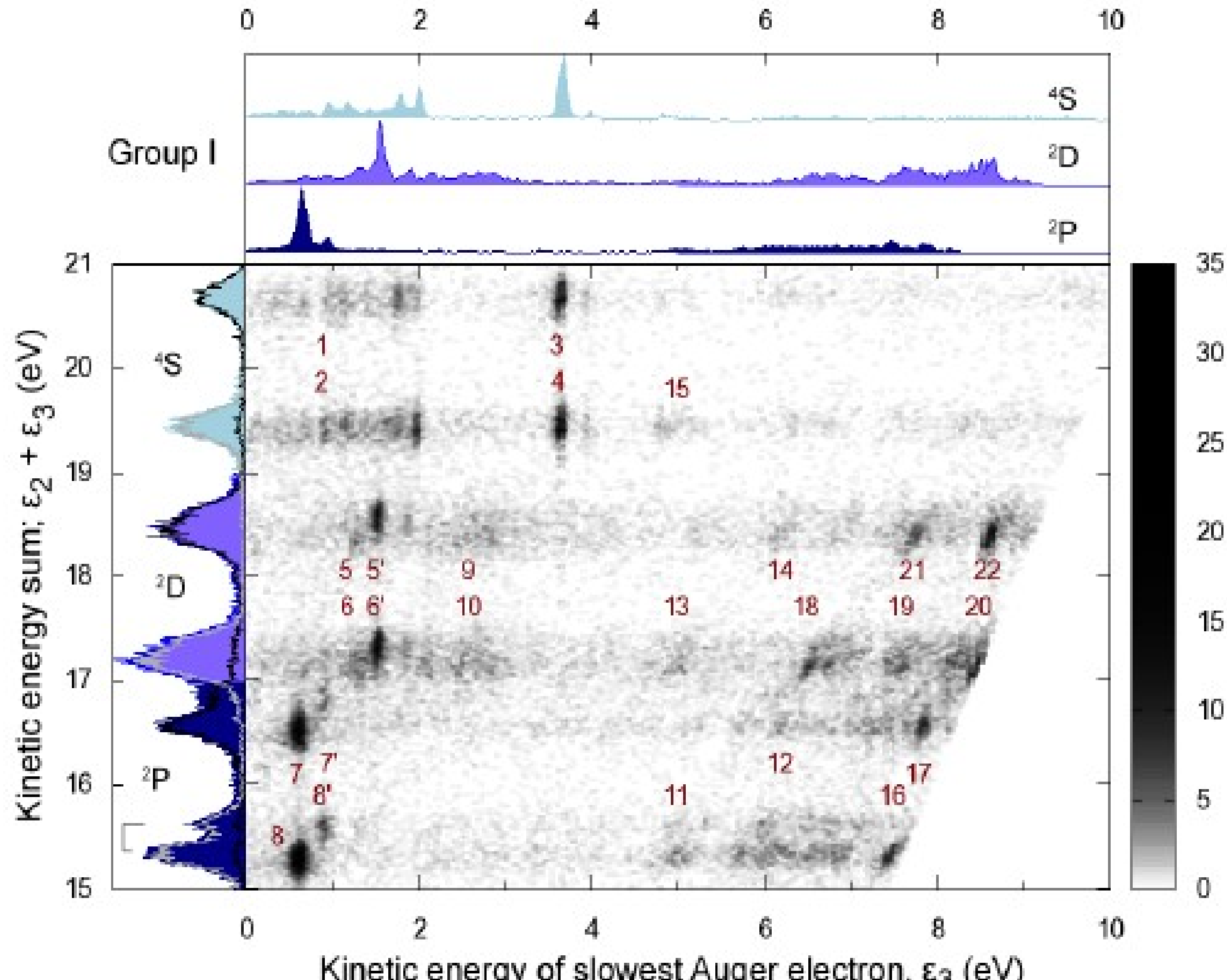
Multiple ionization of noble gases @ synchrotrons

-- coincidence techniques using a magnetic bottle

$Kr \rightarrow Kr^{3+}$

Double Auger decay of 3d-ionized krypton

- Coincidence on 3d photo electron as first arrival electron.
- Six stripes arise from combination of 3d hole states and the ^4S, ^2D and ^2P finals states of Kr^{3+} $4p^{-3}$
- Dark spots refer to Auger lines.



E. Andersson et al, PRA 82 (2010) 043418.

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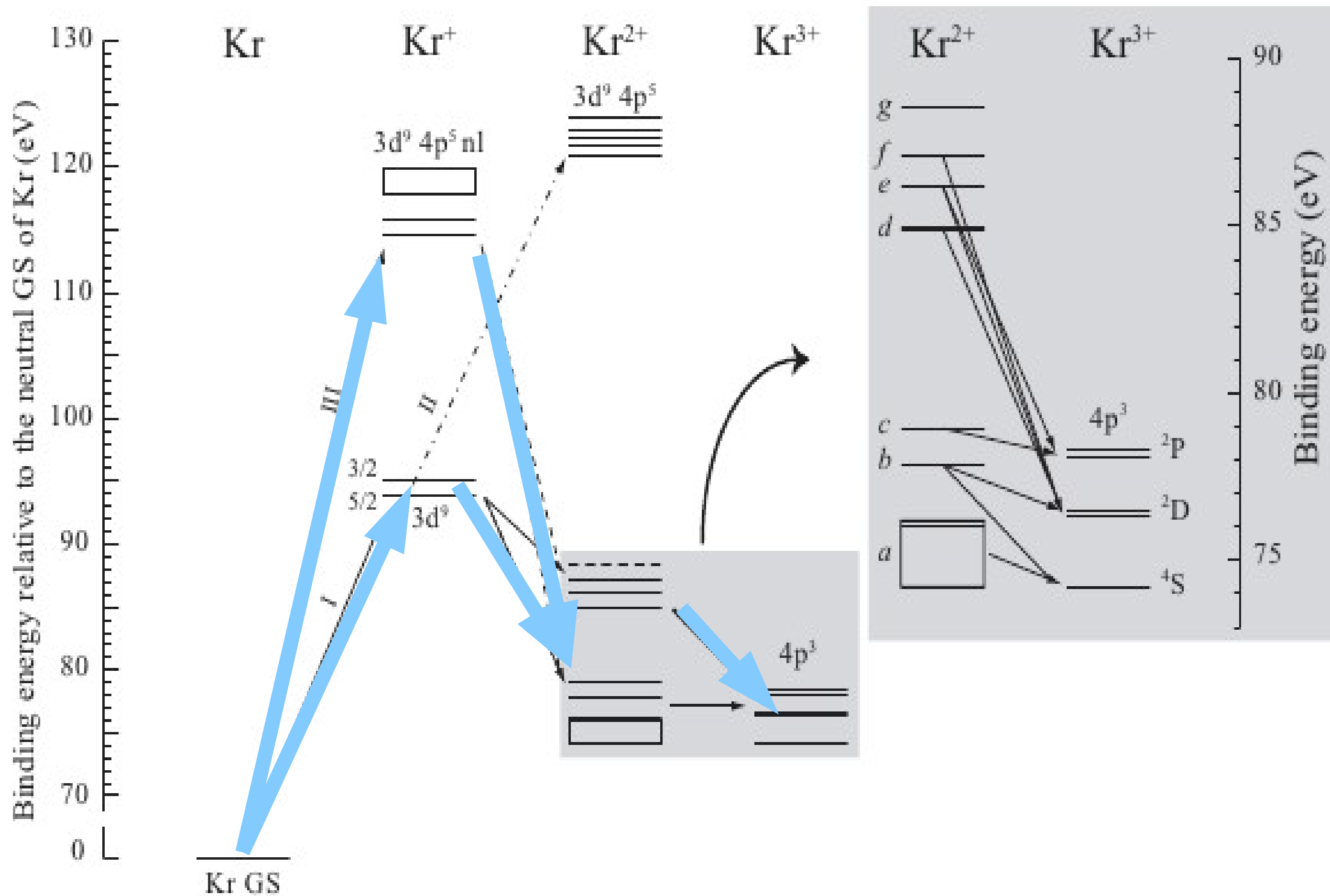


E. Andersson et al, PRA 82 (2010) 043418.

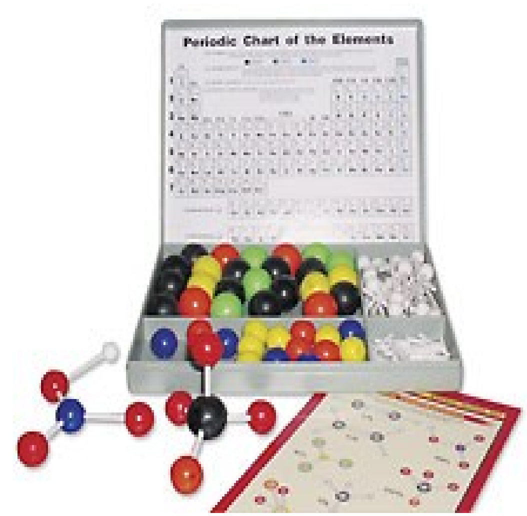
Different `systematic' approaches exist

– to describe the electronic structure of atoms and ions

Multiconfiguration expansions

$$\psi_{\alpha}(P J M) = \sum_{r}^{n_c} c_r(\alpha)\, \gamma_r\, P J M>$$

Construct a `physically motivated' basis in the N-electron Hilbert space.



Many-particle character „electronic correlations“ ⟷ Relativistic effects

Shell structure
static vs. dynamic correlations

Direct vs. indirect effects
QED corrections

Generalization of the knowledge about (Dirac's) one- or few-electron atoms in such a way to enable the „computation“ of heavy atoms and ions.

Multiple ionization of noble gases @ synchrotrons

-- coincidence techniques using a magnetic bottle

Kr → Kr^{3+}

Double Auger decay of 3d-ionized krypton

- Coincidence on 3d photo electron as first arrival electron.
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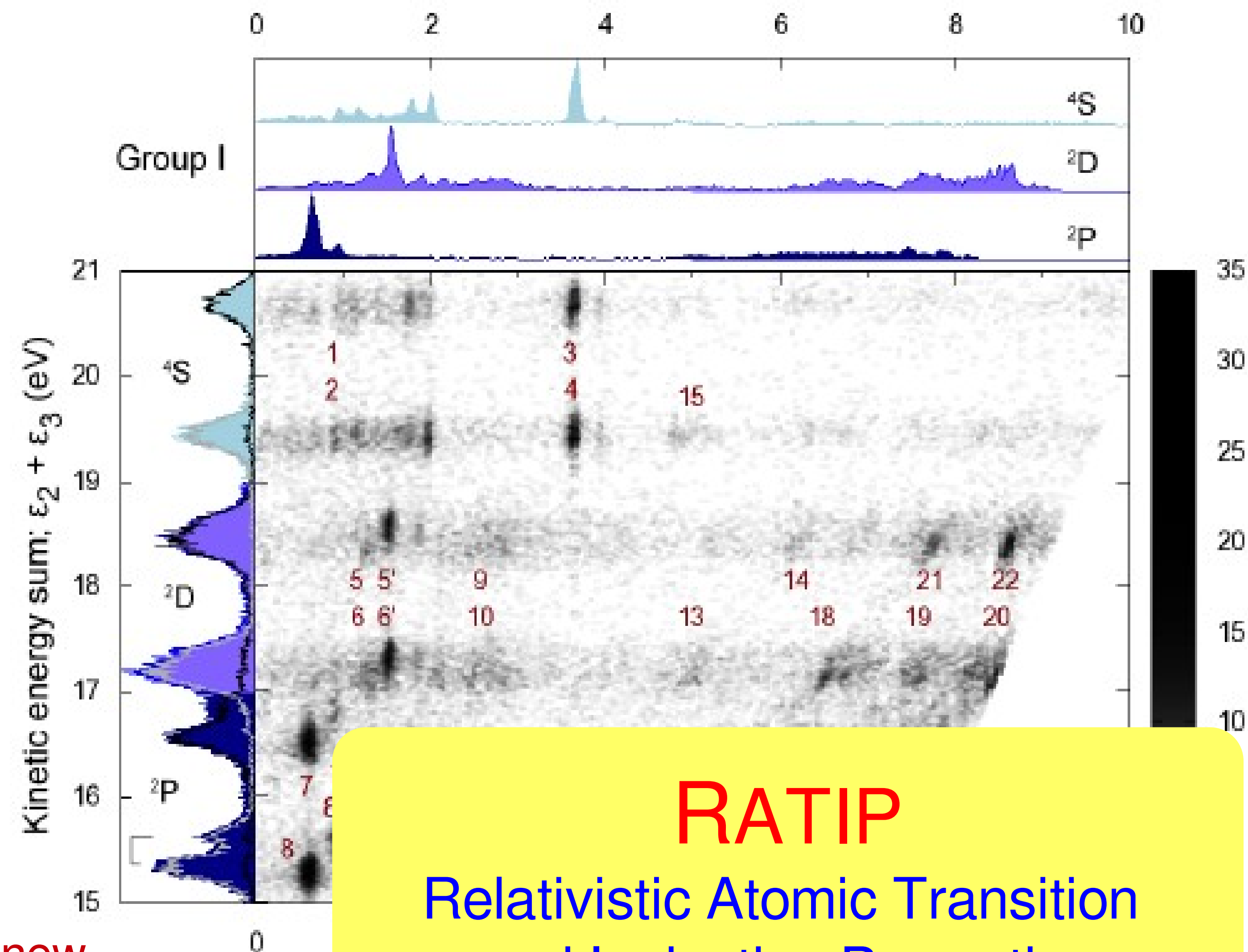


Calculations helped identify new levels and decay pathes in the sequential (auto-) ionization.

RATIP

Relativistic Atomic Transition and Ionization Properties

CPC 183 (2012) 1525 & CPC library

S. Fritzsche, JESRP 114-116 (2001) 1155; Comput. Phys. Commun. 183 (2012) 1525

RATIP

Relativistic Atomic Transition, Ionization and Recombination Properties

AUGER: Auger rates, relative intensities, angular distribution & spin polarization parameters.

REC: Radiative recombination & electron capture rates, angular parameters.

CESD: Determinant representation of atomic and configuration state functions.

RELCI: Relativistic configuration interaction wave functions & QED estimates.

EINSTEIN: Einstein A and B coefficients, transition probabiities & radiative lifetimes.

REOS: Relaxed-orbital Einstein A and B coefficients, transition probabilities and lifetimes.

PHOTO: Ionization cross sections, angular & spin-polarization, alignment of photoions.

TOOLBOX: Level energies and notations; manipuations of file interfaces, miscelaneous.

ANCO: Angular coefficients for scalar and non-scalar operators.

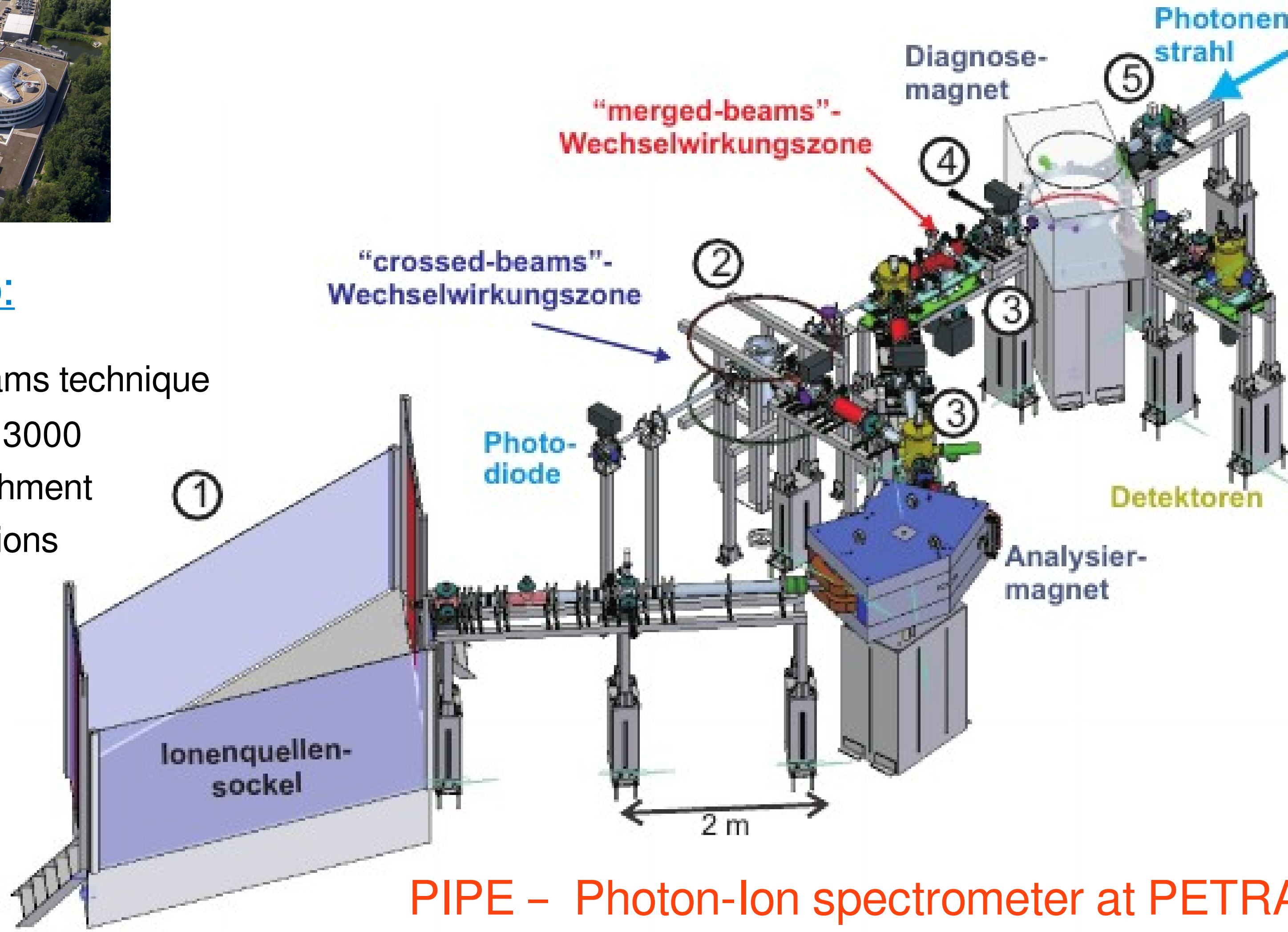
COULOMB: Exitation amplitudes, (M_J-dependent) cross sections, alignment parameters.

Renewed interest on Auger cascades

– PIPE @ soft x-ray beamline of PETRA III

Experimental set-up:

- photon-ion merged-beams technique
- resolving power up to 13000
- double and triple detachment
- widths, branching fractions



PIPE – Photon-Ion spectrometer at PETRA III (Hamburg)

S. Schippers and coworkers, Gießen, Frankfurt, Hamburg collaboration (2015).

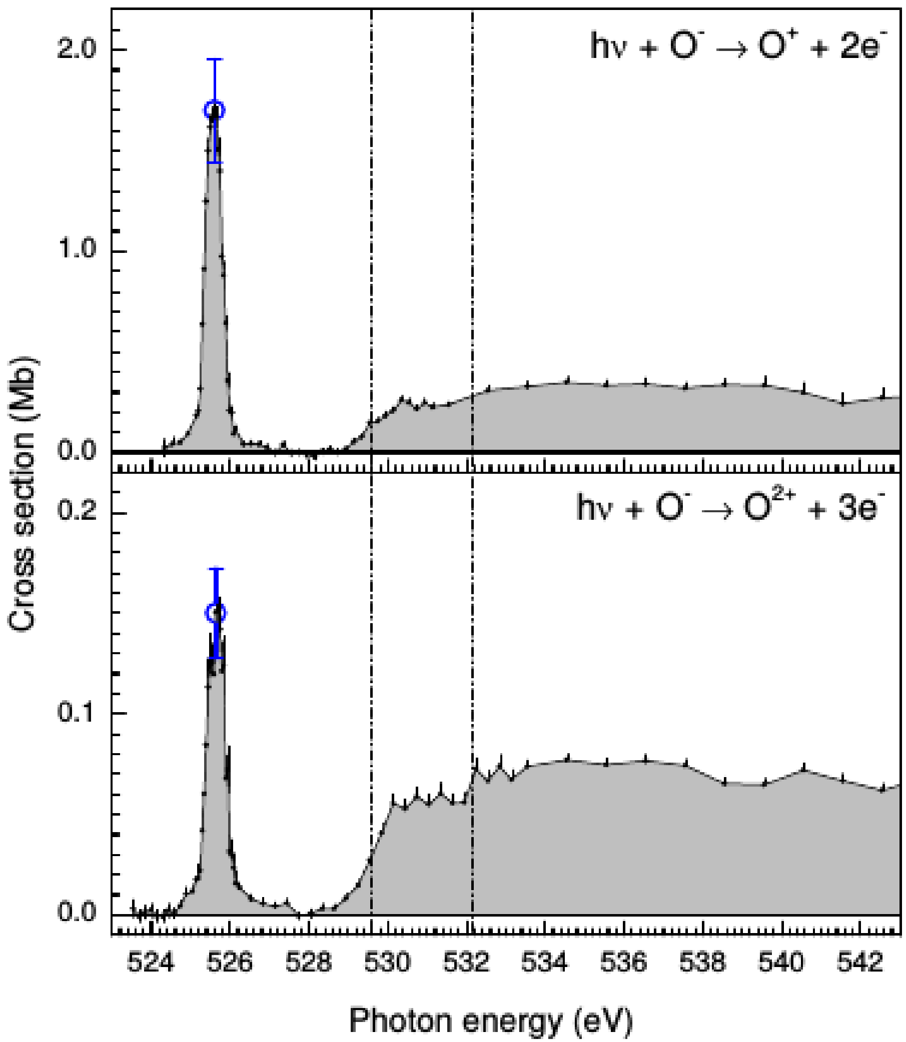
Double & triple photodetachment of oxygen anions

-- exploring the $1s^{-1} \rightarrow 2p^6$ resonance

$$O^- (1s^2\, 2s^2\, 2p^5) + \gamma \rightarrow O^-(1s\, 2s^2\, 2p^6\ {}^2S_{1/2}) \rightarrow O^{(m-1)+} (1s^2\, 2l^{7-m}) + m\, e^-$$

$O^- (1s^{-1} \rightarrow 2p^6)$ resonance:

Quantity	Computed	Exp.
Res. Energy [eV]	525.3	525.6
Ion. Pot. [eV] $^3P_1$	528.4	529.6
$^1P_1$	531.2	532.1
Width [meV]	166	164(14)



S. Schippers et al, PRA 94 (2016) in print.

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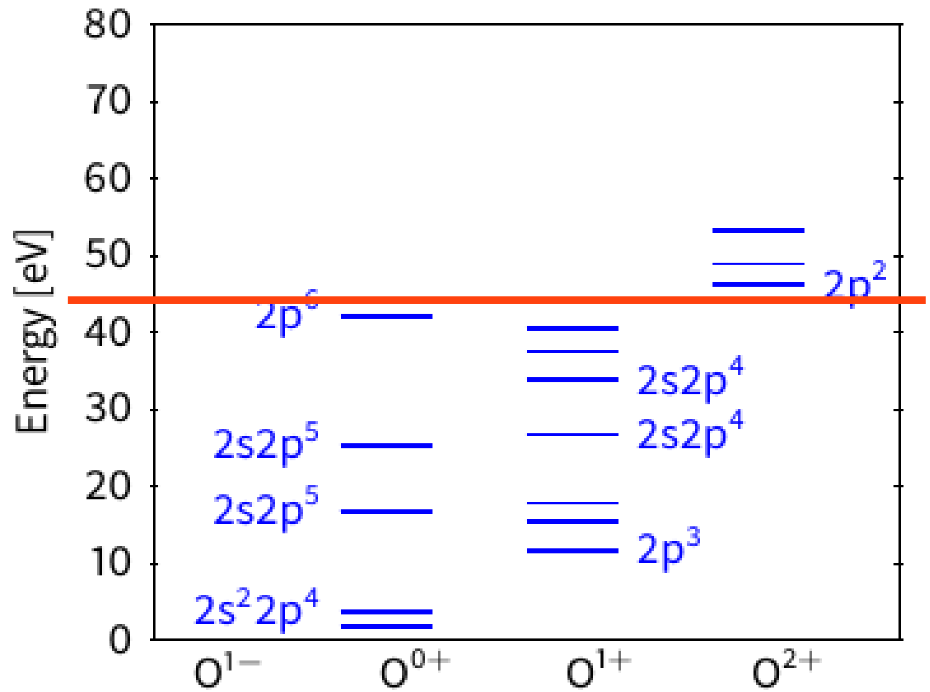
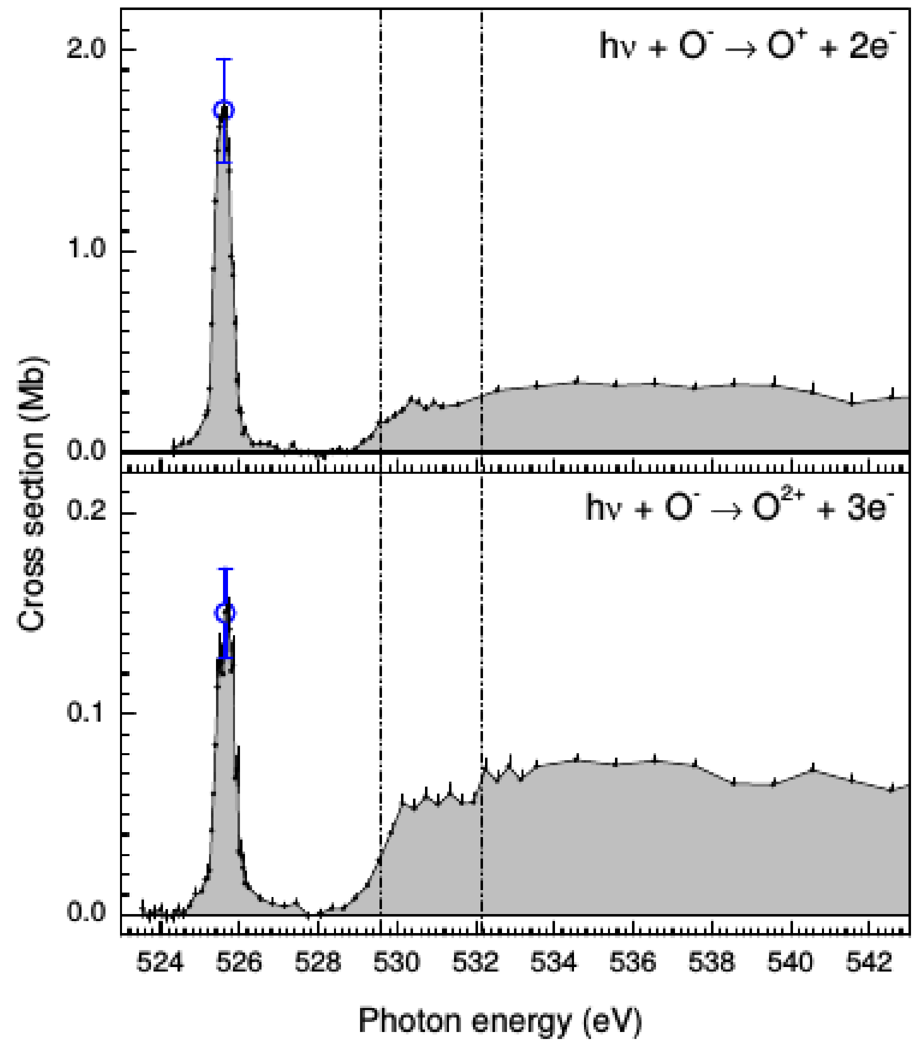


Double & triple detachments by Auger electron emission are energetically forbidden.



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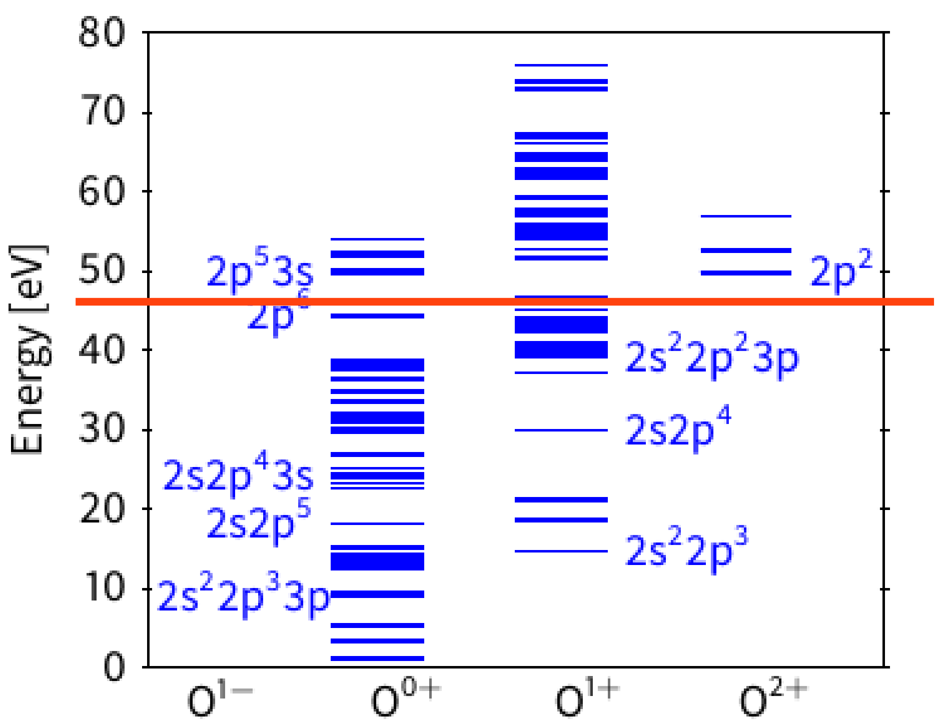
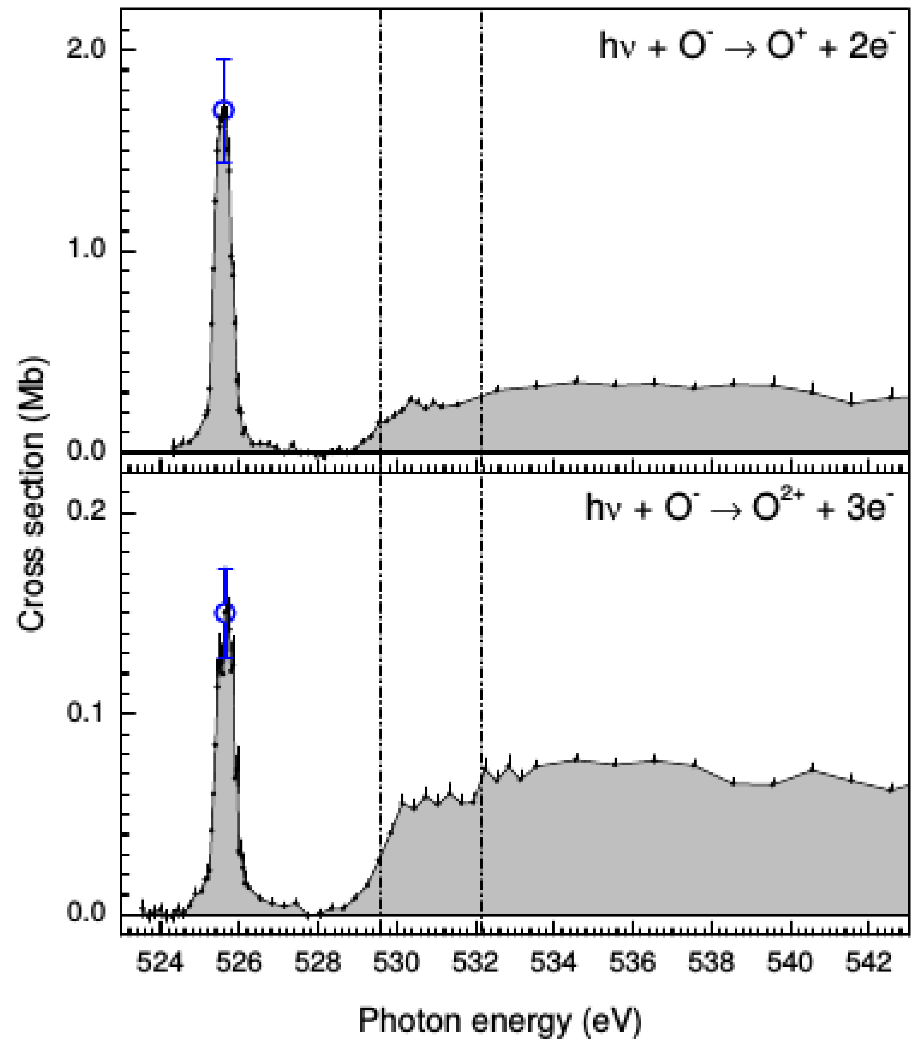




Double & triple detachments by Auger electron emission are energetically forbidden.

→ possible only due to shake-up

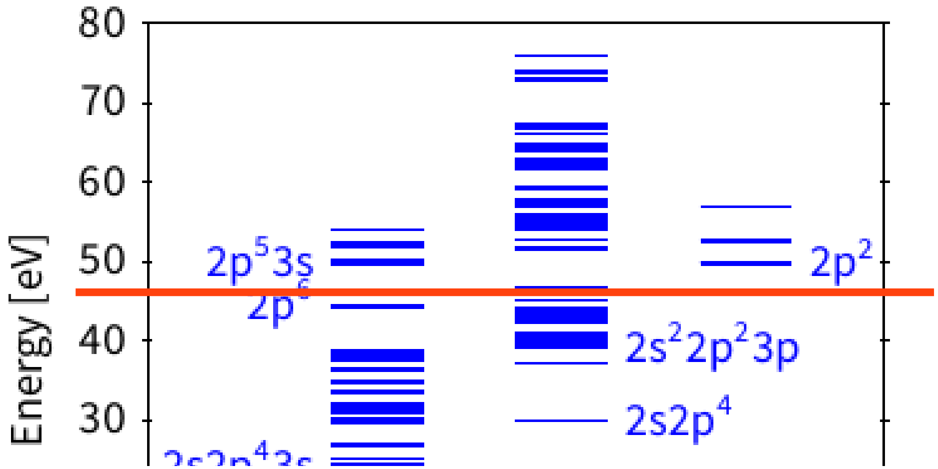
2s → 3s, 2p → 3p

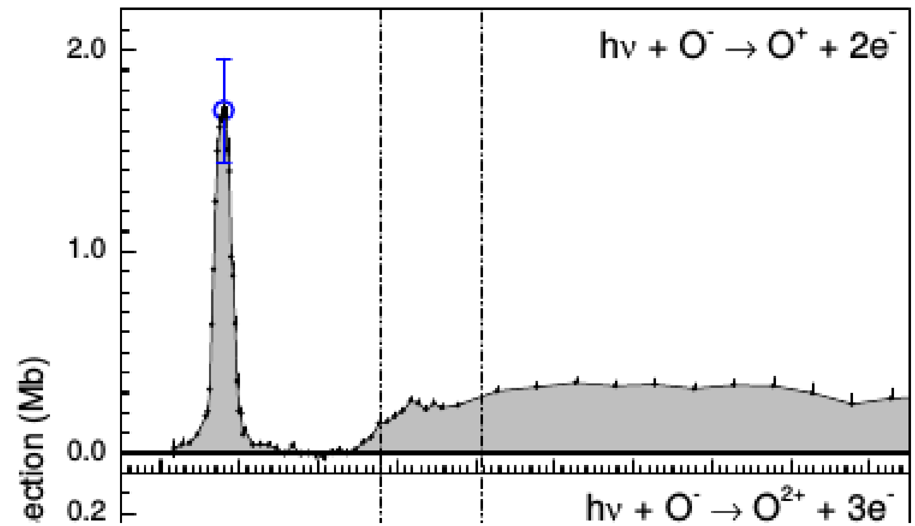
S. Schippers et al, PRA 94 (2016) in print.

Double & triple photodetachment of oxygen anions

-- exploring the $1s^{-1} \rightarrow 2p^6$ resonance

$$O^- (1s^2\, 2s^2\, 2p^5) + \gamma \rightarrow O^-(1s\, 2s^2\, 2p^6\ {}^2S_{1/2}) \rightarrow O^{(m-1)+} (1s^2\, 2l^{7-m}) + m\, e^-$$





Model	Width [meV]	Branching Fraction O	Branching Fraction O^+	Branching Fraction O^{2+}	Ratio O^+/O^{2+}
Simple	133	0.77	0.23	0.0	—
$2s \rightarrow 3s + 2p \rightarrow 3p$	131	0.78	0.22	~ 0.0	—
exp	164 ± 14	—	—	—	10.3

Double & triple photodetachment of oxygen anions

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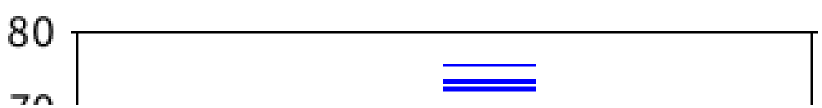

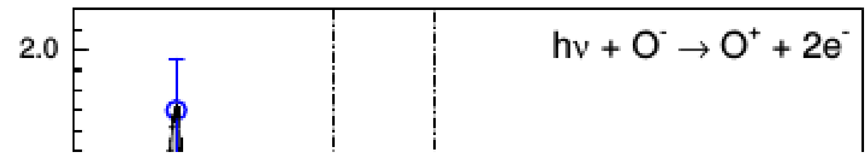

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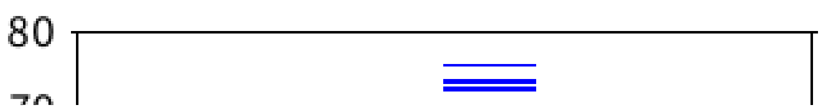
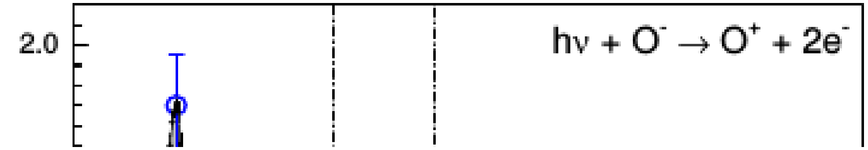
$2s \rightarrow 3s + 2p \rightarrow 3p$	131	0.78	0.22	~ 0.0	—
$+ 2p^2 \rightarrow 3s^2 + 2p^2 \rightarrow 3p^2$	153	0.64	0.36	0.004	106
$+ 2s^2 \rightarrow 3s^2$	161	0.56	0.42	0.016	26
$+ 2s^2 \rightarrow 3p^2$	174	0.46	0.48	0.059	8.1
$+ 2p^2 \rightarrow 3d^2$	166	0.51	0.44	0.042	10.6
exp	164 ± 14	—	—	—	10.3

Double & triple photodetachment of oxygen anions

-- exploring the $1s^{-1} \rightarrow 2p^6$ resonance

$$O^- (1s^2\, 2s^2\, 2p^5) + \gamma \rightarrow O^- (1s\, 2s^2\, 2p^6\ {}^2S_{1/2}) \rightarrow O^{(m-1)+} (1s^2\, 2l^{7-m}) + m\, e^-$$





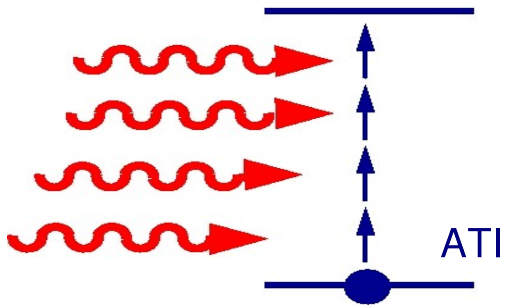
	Width	Branching Fraction			Ratio
Model	[meV]	O			O^+/O^{2+}
Simple	13[illegible]				—
$2s \rightarrow 3s + 2p$ [illegible]					—
$+ 2p^2$ [illegible]					[illegible]06
				[illegible].016	26
			0.48	0.059	8.1
		0.51	0.44	0.042	10.6
	164 ± 14	—	—	—	10.3

Correlated quantum dynamics in weak radiation fields

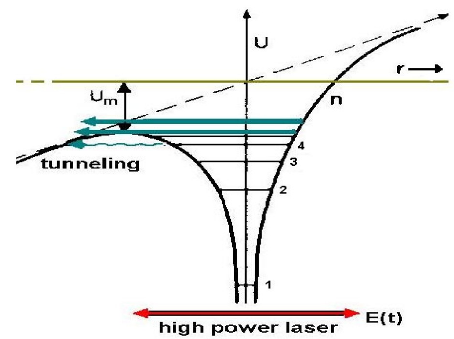
II. Light-matter interactions in intense fields

– from weak- to strong-field ionization

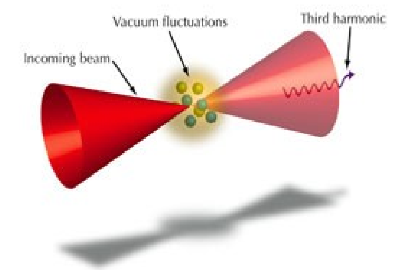
increasing field strength (intensity)



Multiphoton ionization



tunnel ionization



HH and pairs from vacuum

increasing frequency (photon energy)

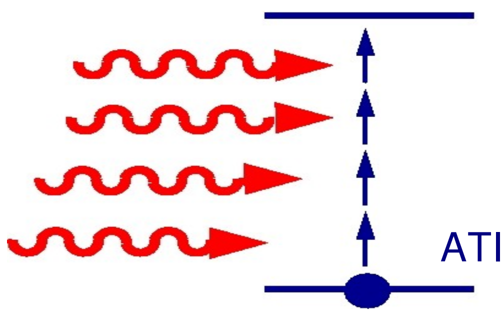
- Excitation & ionization at (ultra-) fast time scales & relativistic photon energies
- Electron dynamics in intense FEL radiation

  (multi-photon & multi-color ionization; coherently driven dynamics of inner-shell excited; sidebands; decoherence & quantum beats, ...)
- Creation and dynamics of warm dense matter

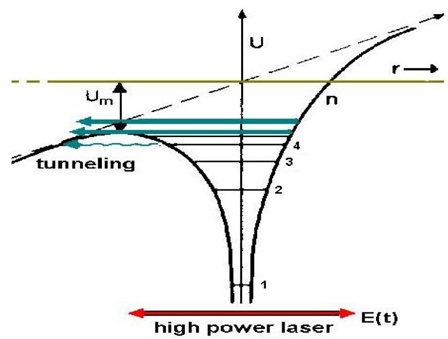
Light-matter interactions in intense fields

– from weak- to strong-field ionization

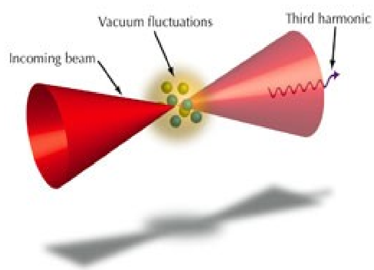
increasing field strength (intensity)



Multiphoton ionization



tunnel ionization



HH and pairs from vacuum

increasing frequency (photon energy)

- Excitation & ionization at (ultra-) fast tim
- Electron dynamics in intense FEL radiati

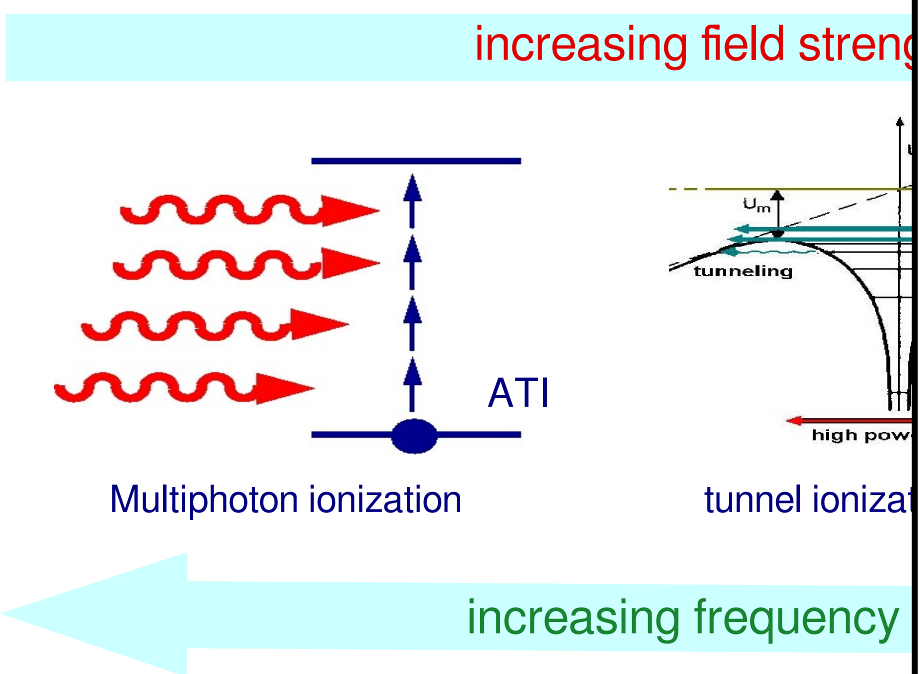
  (multi-photon & multi-color ionization; coher
  sidebands; decoherence & quantum beats, ...)
- Creation and dynamics of warm dense n

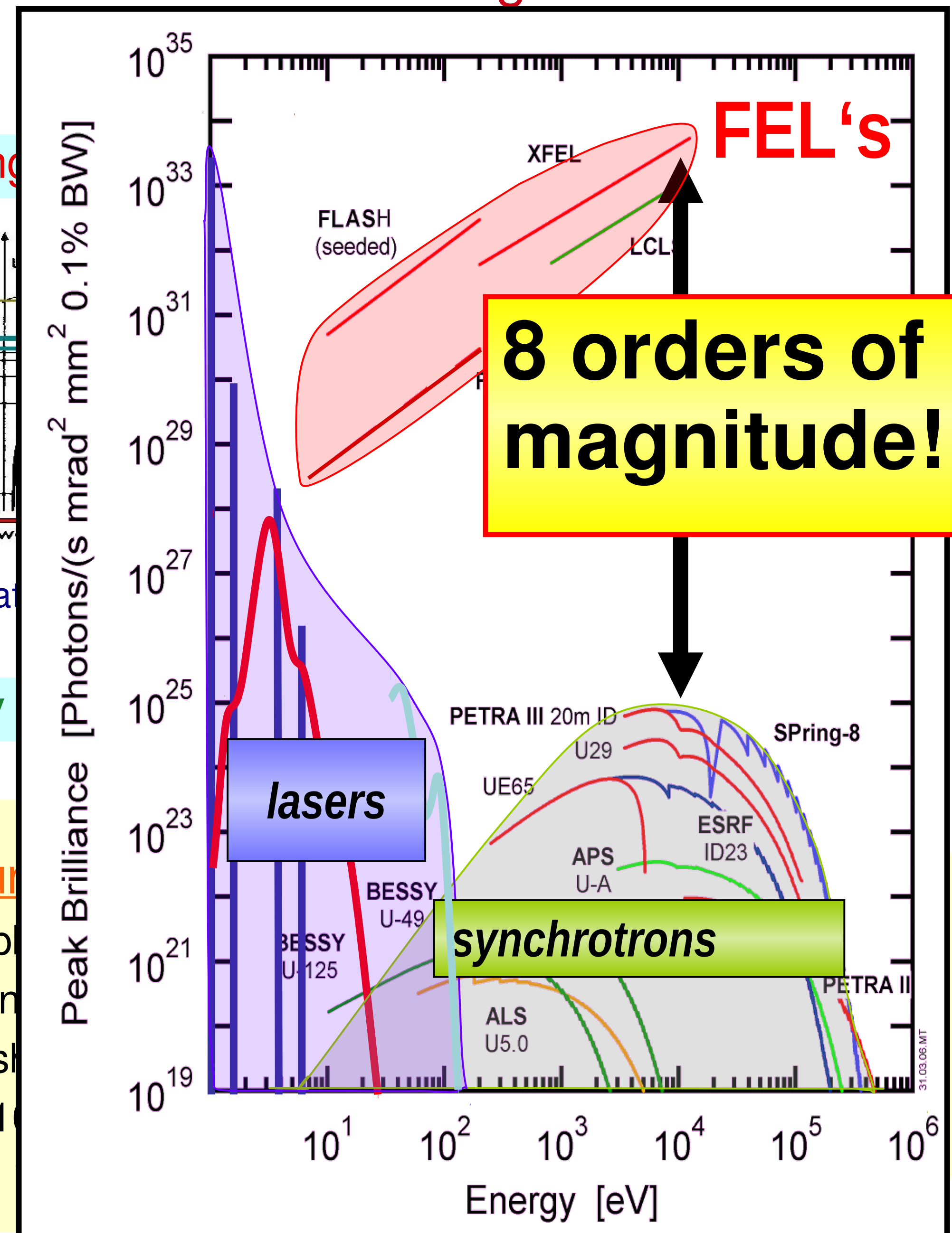
Current FEL parameters:

- photon energies 10 eV – 10 keV
- intensities 10^{13} -10^{16} W/cm^2
- short pulses (few - 50 fs)
- 10^6 -10^8 photons / target atom
  → nonlinear processes in the VUV & x-ray region

Light-matter interactions in intense fields

– from weak- to strong-field ionization





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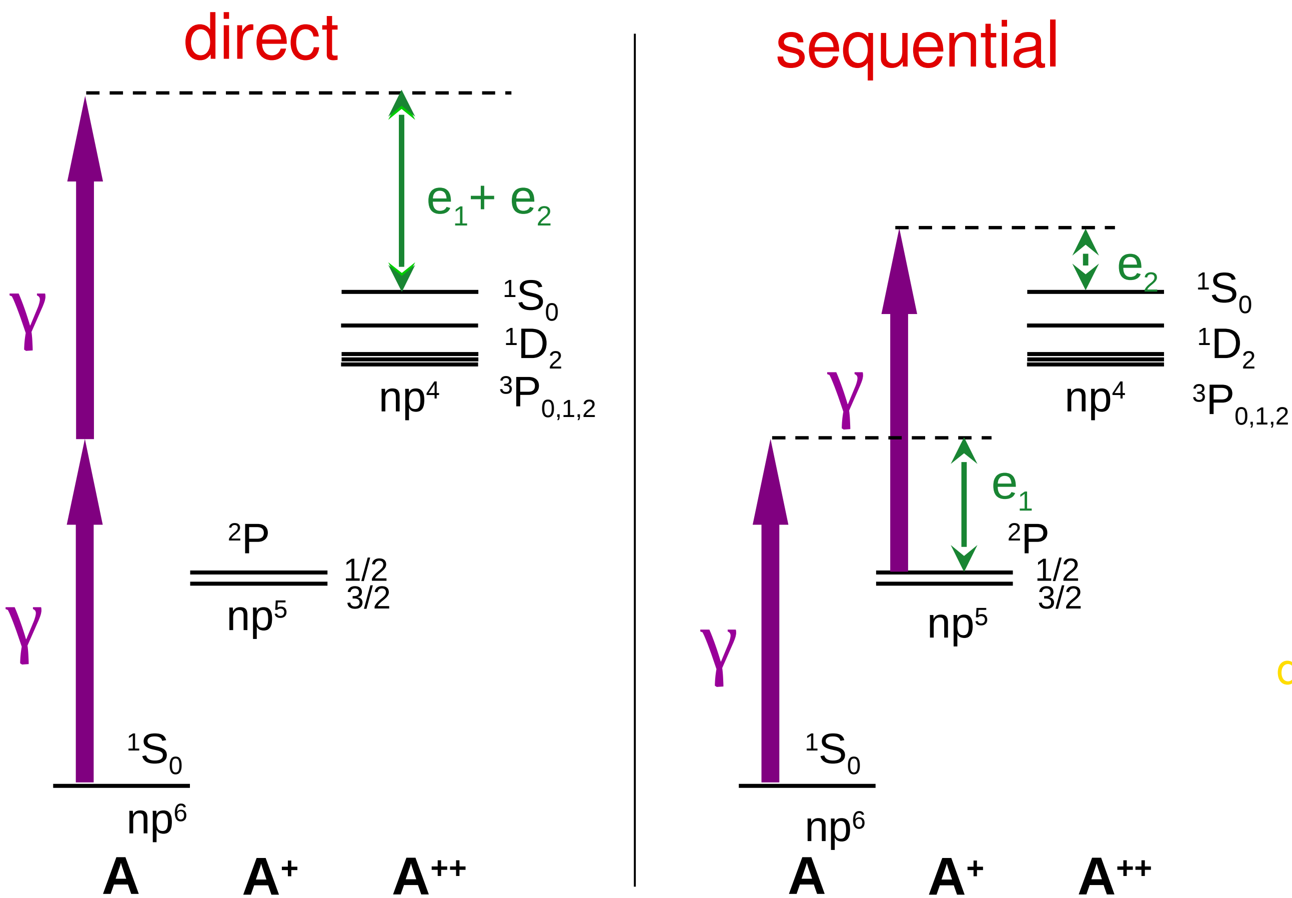
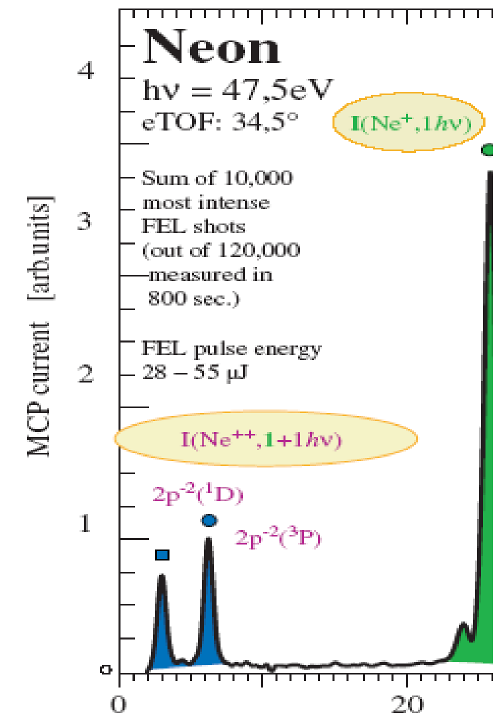
Two-photon double ionization (TPDI)

-- sequential vs. direct knockout of several electrons

How differs the (dynamical) light-matter coupling via real and `virtual' intermediate states ?





M. Braune et al, ICPEAC (2007/2009).

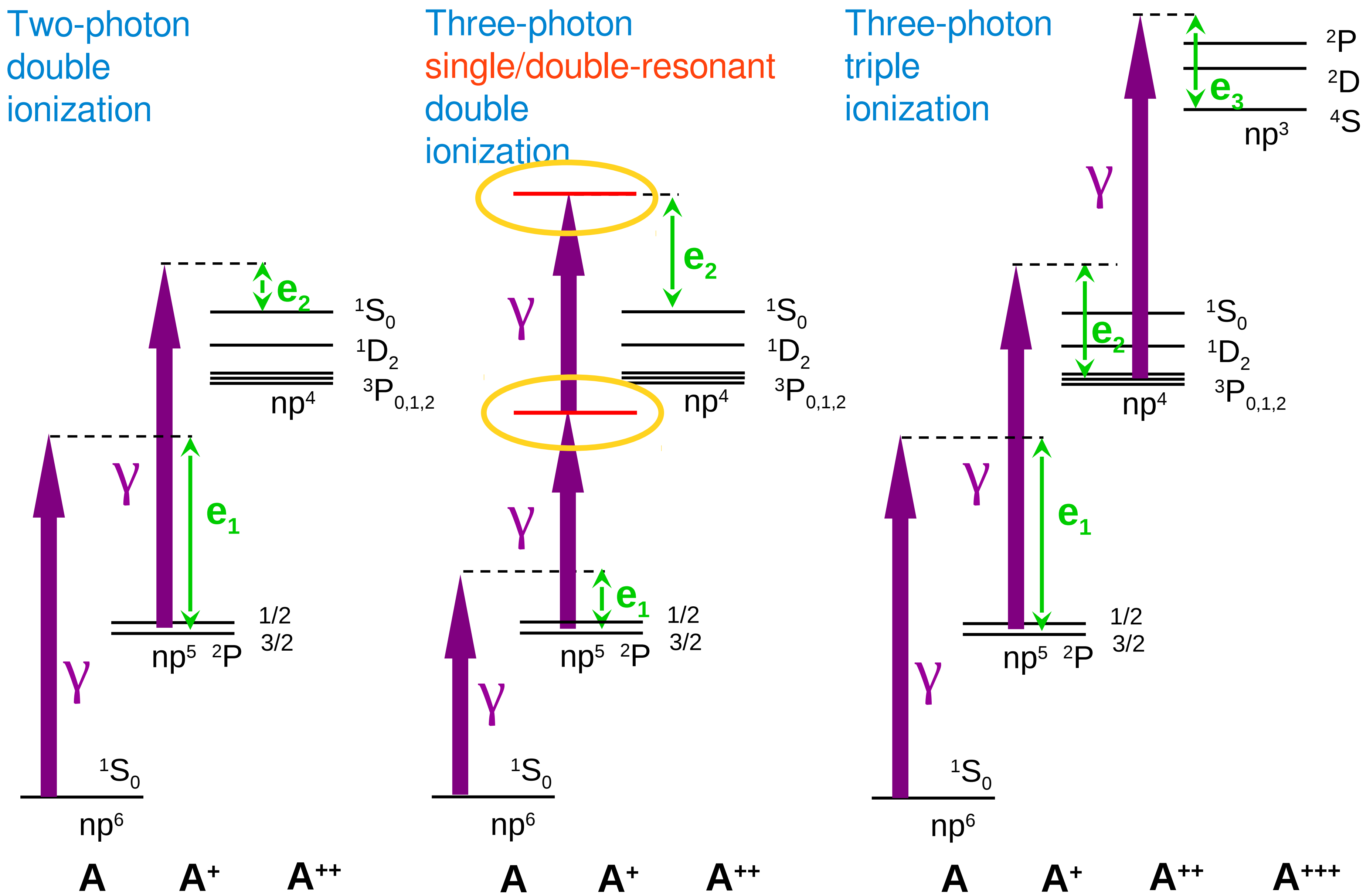
$$\frac{dN^{+}}{dt} = \frac{N}{T}\,\sigma^{(0\to 1)}\left(\frac{\mathcal{N}_{\rm ph}}{A}\right)$$

dynamical coupling ?

$$\frac{dN^{2+}}{dt} = \frac{N}{T}\,\sigma^{(0\to 2)}\left(\frac{\mathcal{N}_{\rm ph}}{A}\right)^{2}$$

Further two- and three-photon processes

-- sequential vs. direct ionization



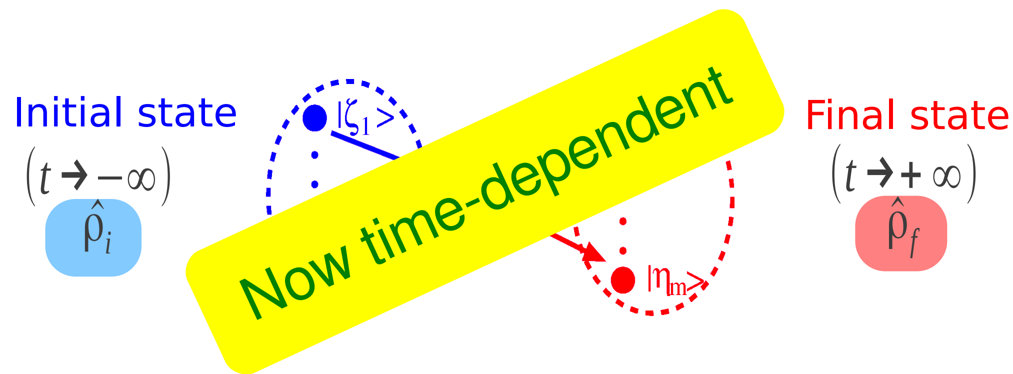
Coherent time evolution of inner-shell excited systems

– in short-pulse or pump-probe experiments

Explicitly time-dependent density operator including spatial degrees of freedom:

$$\dot{\rho} = \frac{i}{\hbar}\,[H,\,\rho] + L\,\rho$$

Based Liouville's equation



$$i\,\frac{d\rho_{kq}(\alpha,\beta)}{dt} = \sum_{\kappa' q'} \sum_{\gamma} \Bigg\{ F_{\kappa q}^{\kappa' q'}\left(\alpha,\beta,\gamma, \begin{matrix}\text{pulses}\\ \text{geometry}\end{matrix}; t\right) \rho_{k'q'}(\gamma,\beta)$$

direct coupling

$$- G_{\kappa q}^{\kappa' q'}\left(\alpha,\beta,\gamma, \begin{matrix}\text{pulses}\\ \text{geometry}\end{matrix}; t\right) \rho_{k'q'}(\alpha,\gamma)$$

exchange

$$- i\,\Gamma_{\kappa q}^{\kappa' q'}(\alpha,\beta,\gamma;t)\, \rho_{k'q'}(\gamma,\beta) \Bigg\}$$

ionization & loss processes

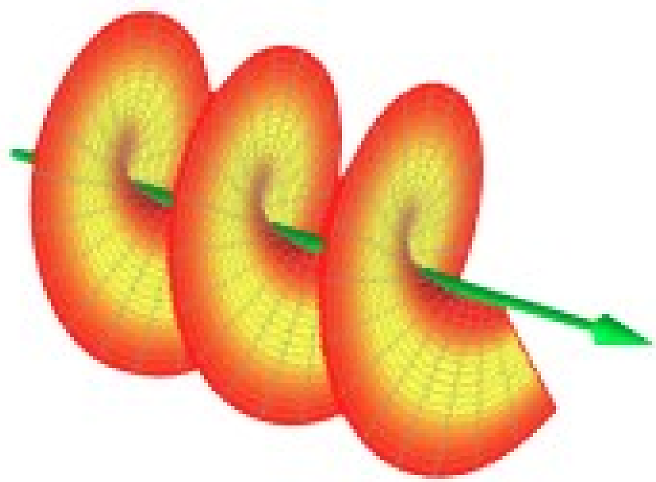
Atomic (transition & ionization) amplitudes from many-body theory (RATIP)

S. Fritzsche, CPC 183 (2012) 1525

Collaboration with Alexei Grum-Grzhimailo

III. Atomic interactions with twisted light and electrons

– waves with helical wave fronts and orbital angular momentum



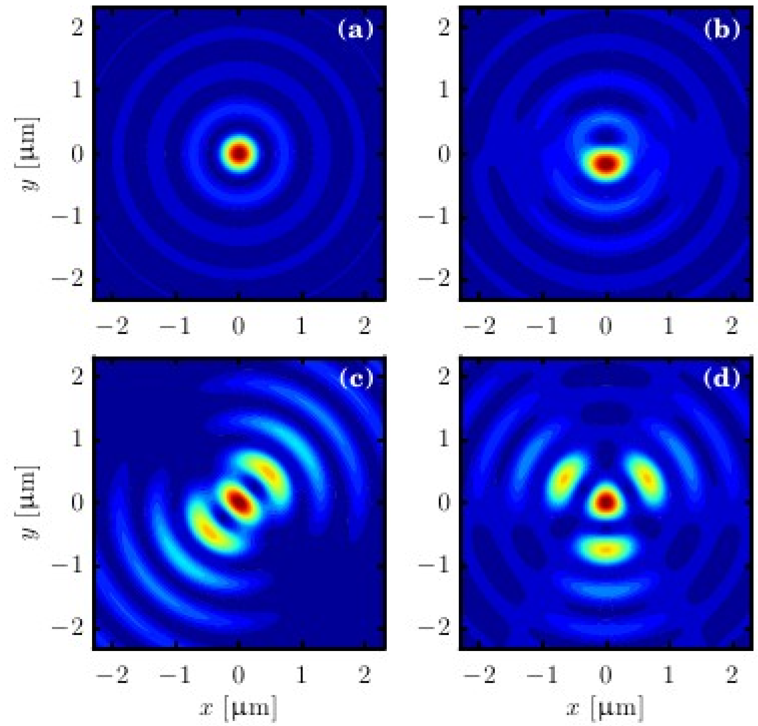
- Laguerre-Gaussian beams
- Bessel beams
- Vector beams

$$\psi \sim e^{-i\omega t + ik_z z}\, e^{im\varphi}\, J_m(k_\perp r)$$

Quantum numbers: k_z, k_{perp}, m, λ

Topological charge, winding number, projection of OAM, ...

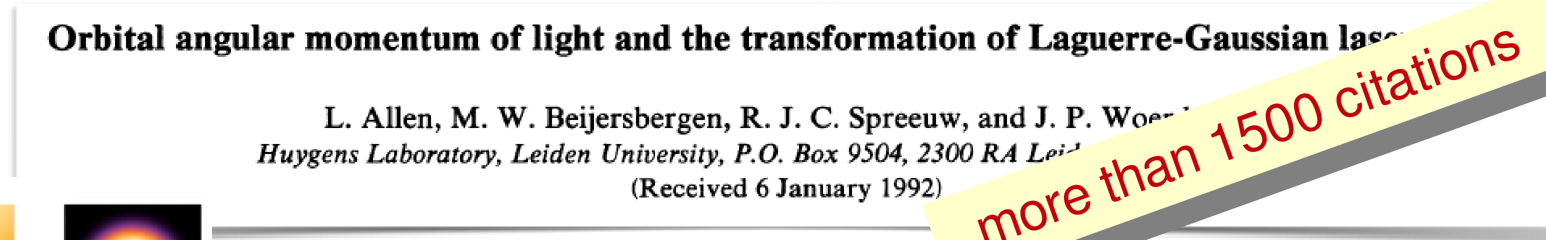
Superposition of Bessel beams



Twisted photons

– beams with vortex lines and spiral phase fronts

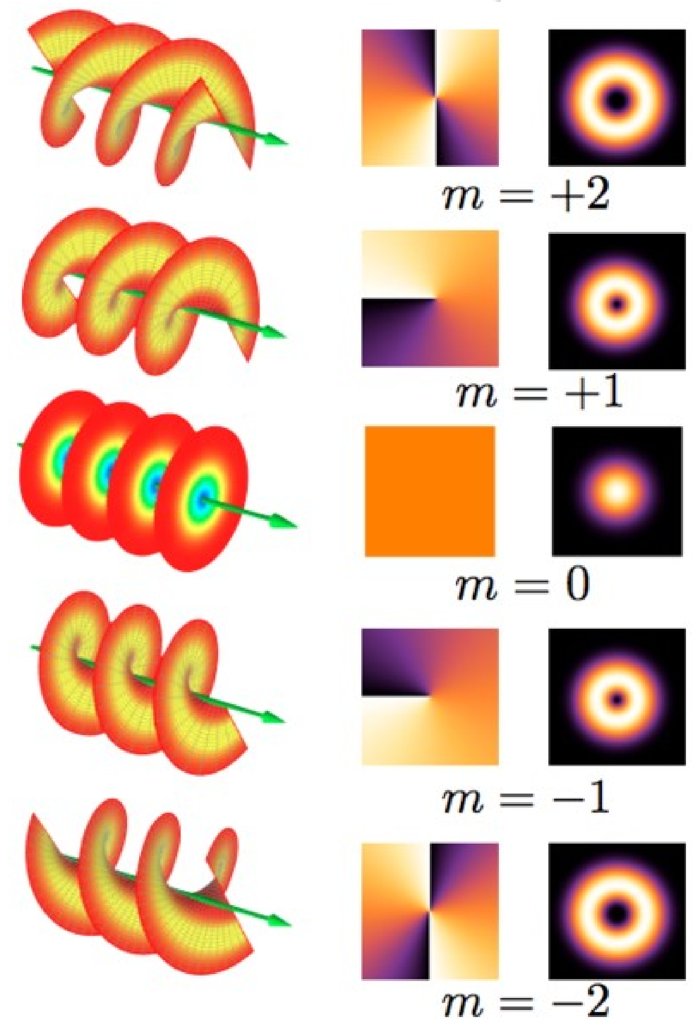
$A \sim \exp(i\, m_{OAM}\, \varphi)$


Orbital angular momentum of light and the transformation of Laguerre-Gaussian las…

L. Allen, M. W. Beijersbergen, R. J. C. Spreeuw, and J. P. Woer…
Huygens Laboratory, Leiden University, P.O. Box 9504, 2300 RA Lei…
(Received 6 January 1992)

more than 1500 citations

PRA **45**, 8185 (1992)



photon spin → polarization

orbital angular momentum → vortex lines & helical phases

New insights into the light-matter interaction, including a good number of novel applications

C. Yao and M. Padgett, Adv. Optics & Photon. 3 (2011) 161.

Solutions of Maxwell's wave equations

– using especially Helmholtz' equation for the spatial part $\psi(\mathbf{r})$

$$\Box\,\psi(\mathbf{r},t) \;=\; \left(\nabla^2 \;-\; \frac{1}{c^2}\frac{\partial^2}{\partial^2\,t^2}\right)\,\psi(\mathbf{r},t) \;=\; 0$$

$$\psi(\mathbf{r},t) \;=\; \psi(\mathbf{r})\,T(t) \qquad\qquad \text{Separation mit Konstante } -k^2$$

$$\boxed{\left(\nabla^2 \;+\; k^2\right)\,\psi(\mathbf{r}) \;=\; 0\,,} \qquad\qquad \left(\frac{d^2}{dt^2} \;+\; \omega^2\right)\,T(t) \;=\; 0\,, \qquad\qquad \omega \;=\; ck$$

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For different coordinates & applications:

(1) Plane waves (sinusoidal, spectral decomposition, cartesian)

(2) Multipole expansions, spherical waves (spherical)

(3) Hermite-Gaussian beams (TEM modes, ...)

(4) Laguerre-Gaussian beams (paraxial, cylindrical)

(5) Vector beams

(6) Bessel beams

$$\boxed{\mathbf{A}^{\mathrm{tw}}(\mathbf{r}) = \int a_{\varkappa m_\gamma}(\mathbf{k}_\perp)\, \mathbf{e}_{\mathbf{k}\Lambda}\, \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{r}}\, \frac{\mathrm{d}^2\mathbf{k}_\perp}{(2\pi)^2}}$$

$\longrightarrow$ and many more that can be realized experimentally today !!

Solutions of Maxwell's wave equations

– using especially Helmholtz' equation [illegible])

$$\Box\, \psi(\mathbf{r}, t) \;=\; \left(\nabla^2 \;-\; \frac{1}{c^2} \frac{\partial^2}{\partial [illegible]} \right) [illegible]$$

$$\psi(\mathbf{r}, t) \;=\; [illegible] \text{stante} \;-\; k^2$$

$$[illegible] \left(\frac{d^2}{dt^2} + \omega^2 \right) T(t) \;=\; 0\,, \qquad \omega \;=\; ck$$

Twisted photons each have a well-defined (projection of orbital) angular momentum; OAM light.

[illegible]or different coordinates & applications:

(1) Plane waves (sinusoidal, spectral decomposition, cartesian)

(2) Multipole expansions, spherical waves (spherical)

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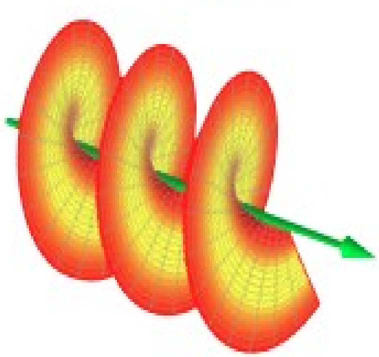
$\longrightarrow$ and many more that can be realized experimentally today !!

Our recent focus: Bessel beams

– with well-defined AM, monochromatic and <u>non-diffractive</u>



$$\psi \sim e^{-i\omega t + ik_z z}\, e^{im\varphi}\, J_m(k_\perp r)$$

Quantum numbers: k_z, k_{perp}, m_γ, λ

Vector potential:

$$\mathbf{A}^{tw}(\mathbf{r}) = \int a_{\varkappa m_\gamma}(\mathbf{k}_\perp)\, \mathbf{e}_{\mathbf{k}\Lambda}\, e^{i\mathbf{k}\cdot\mathbf{r}} \frac{d^2\mathbf{k}_\perp}{(2\pi)^2}$$

Fullfilles Helmholtz's equation.

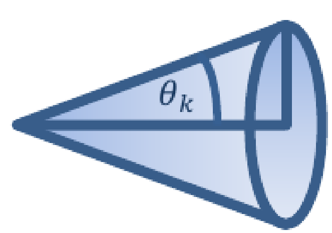
Probabilities of individual OAM components:

$$P_{m_\gamma - 1} = \frac{1}{4}(\cos\theta_k + \lambda)^2$$

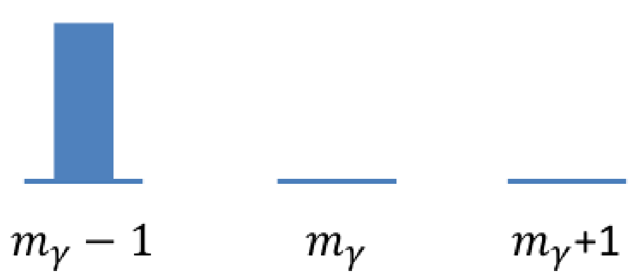
$$P_{m_\gamma} = \frac{1}{2}(\sin\theta_k)^2$$

$$P_{m_\gamma + 1} = \frac{1}{4}(\cos\theta_k - \lambda)^2$$

Bessel beam with small opening angle θ_k

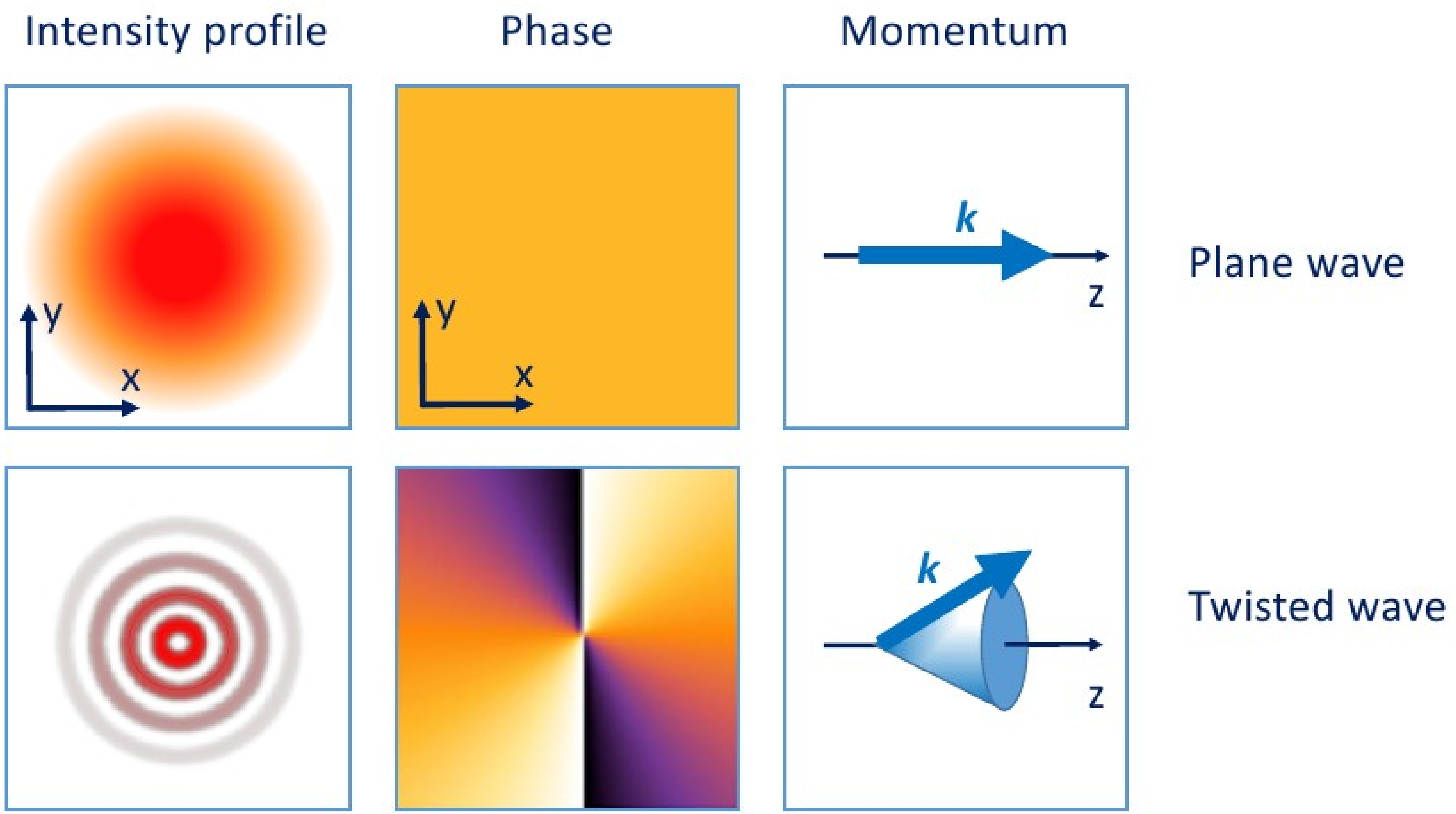


$\theta_k = 1°, \lambda = +1$ ~ left circular



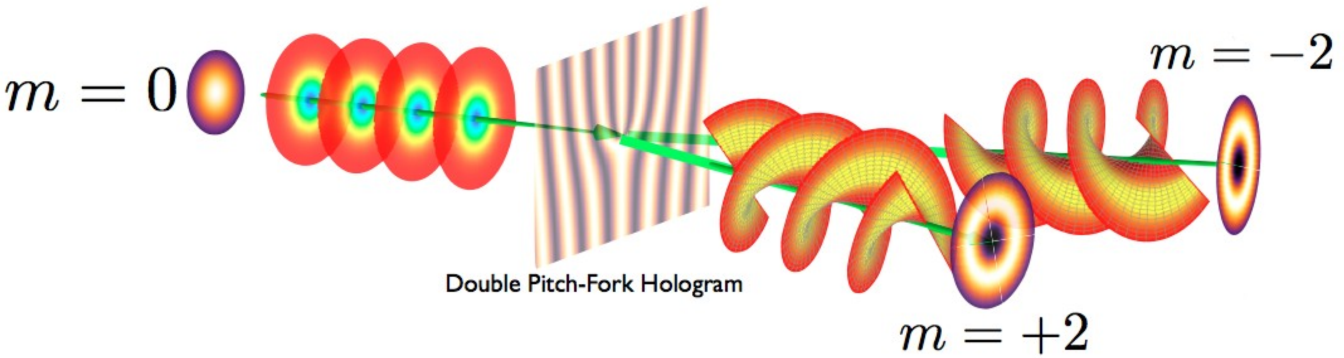
Bessel beams *vs.* plane waves

– Representation in position, phase and momentum



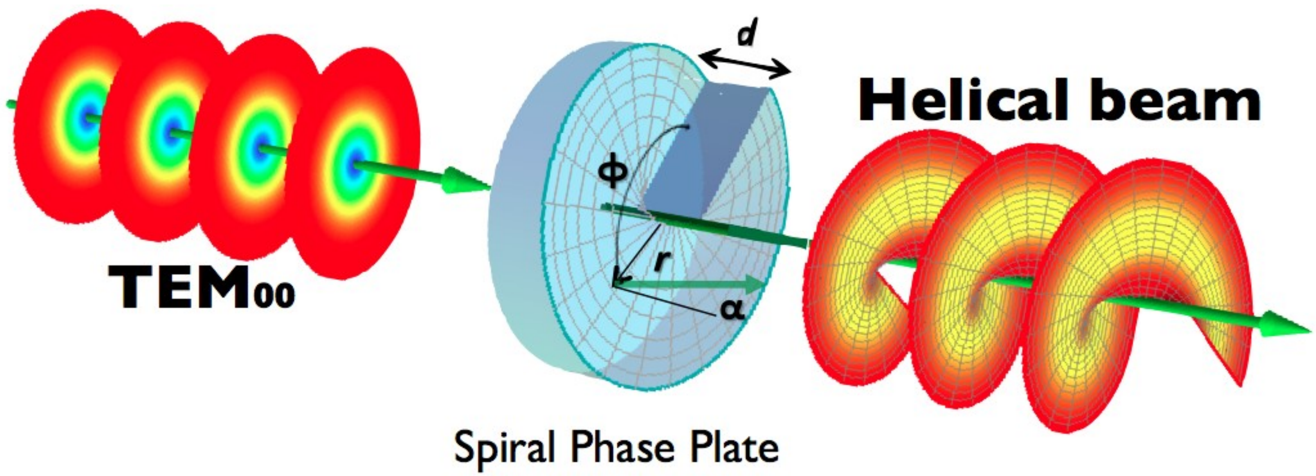
Generation of twisted photons

– due to different experimental techniques



pitch-fork hologram

spiral phase plate

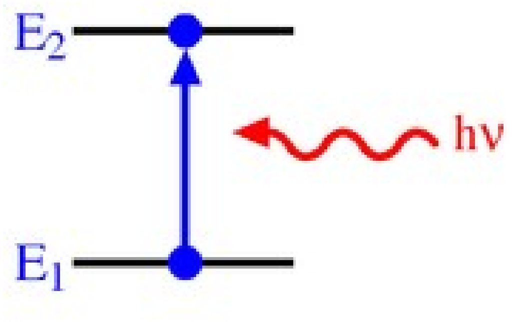


... as well as by several other methods, such as q-plates, spatial & cylindrical mode converters, axicons and also integrated circuits.

C. Yao and M. Padgett, Adv. Optics & Photon. 3 (2011) 161.

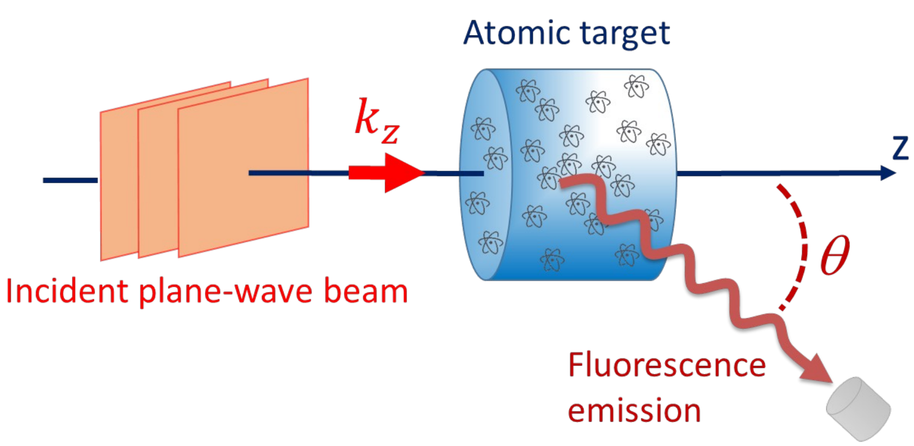
Photoabsorption and photoionization of light

– understanding basic light-matter interactions

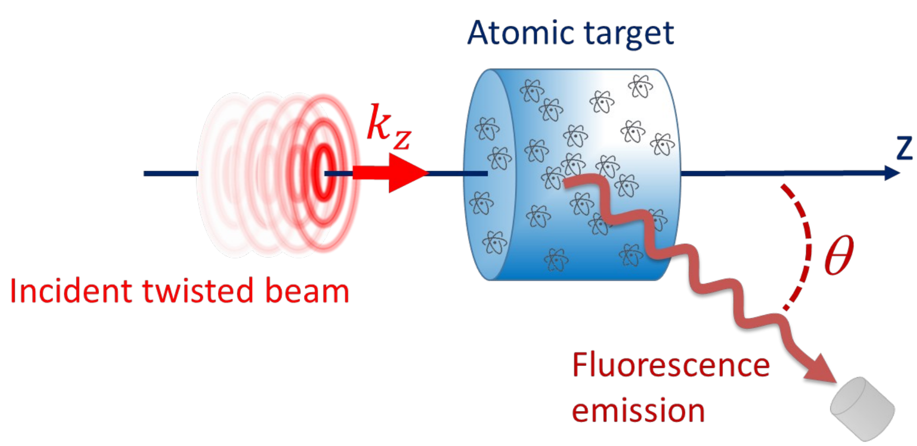


- photochemistry
- x-ray source
- optically pumped lasers
- solar cells
- photochromic complexes

Plane-wave light



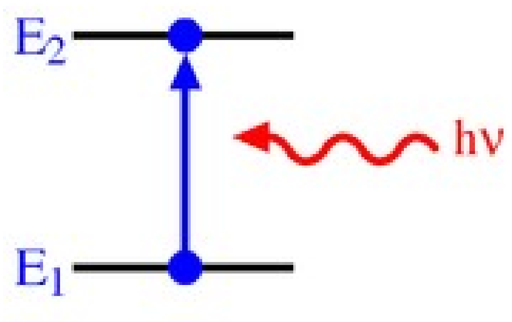
Twisted light



How differs the photo-absorption and ionization for plane-wave & twisted radiation ?

Photoabsorption and photoionization of light

– understanding basic light-matter interactions



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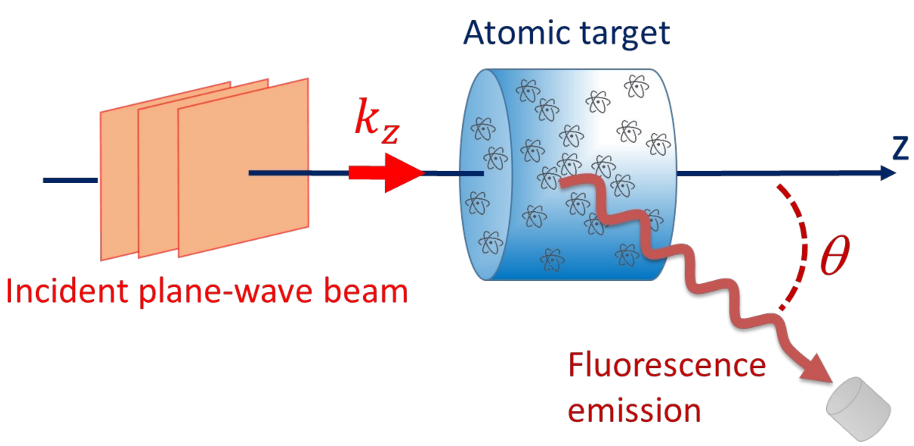
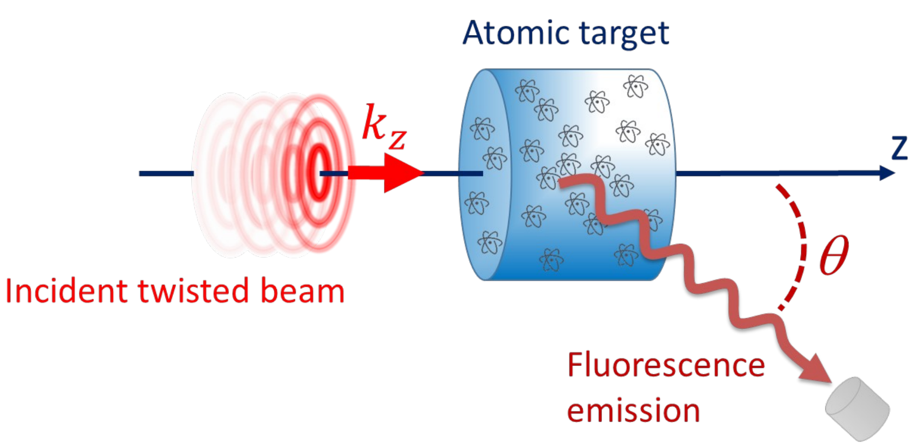
Plane-wave light



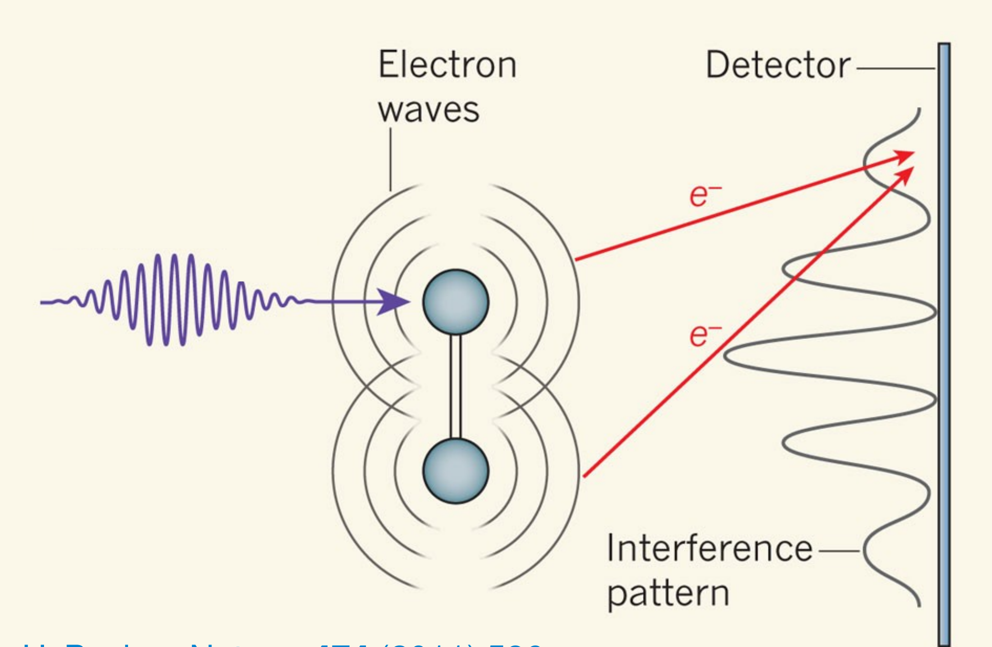
Twisted light



How differs the photo-absorption and ionization for plane-wave & twisted radiation ?

Photoionization of diatomic molecules

– the atomic version of Youngs' double-slit experiment



U. Becker, Natrure **474** (2011) 586.

Scattering of light at the (two) atomic centers leads to interferences in the observed cross sections.

Fano's model:

$$\frac{d\sigma}{d\Omega_f} \sim \frac{1}{\omega^2}\, cos^2(\boldsymbol{k}_f \cdot \boldsymbol{R}/2)$$

Photoionization of diatomic molecules

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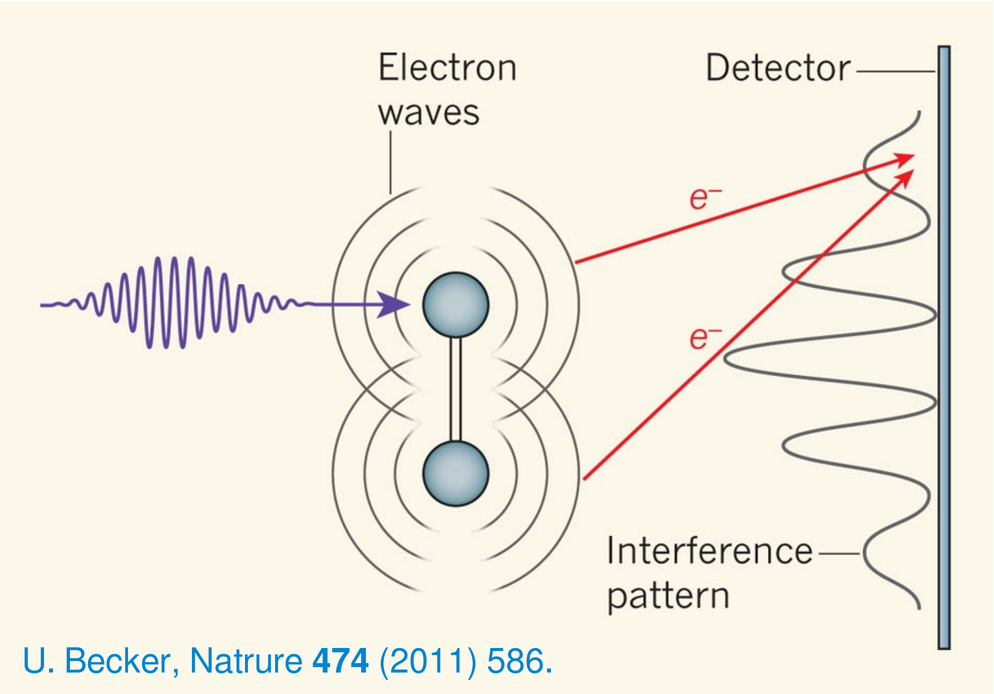

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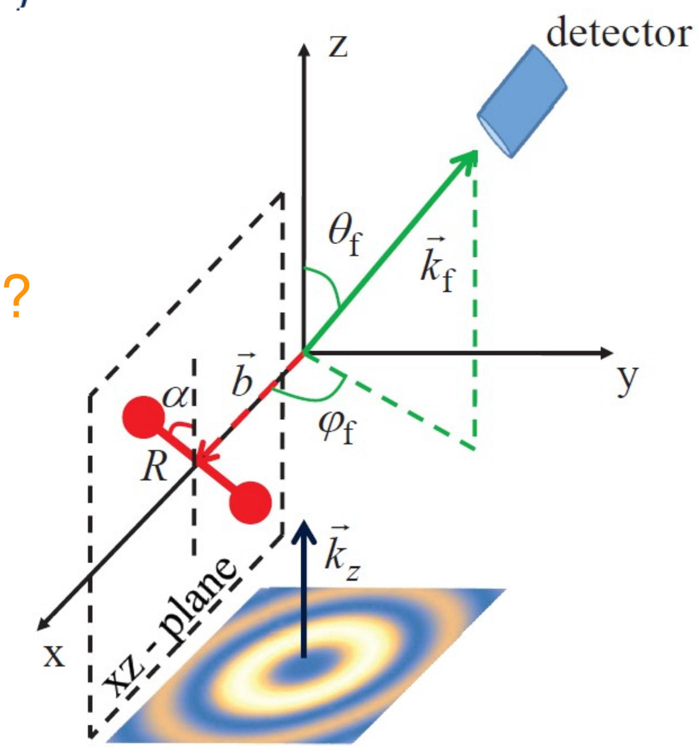


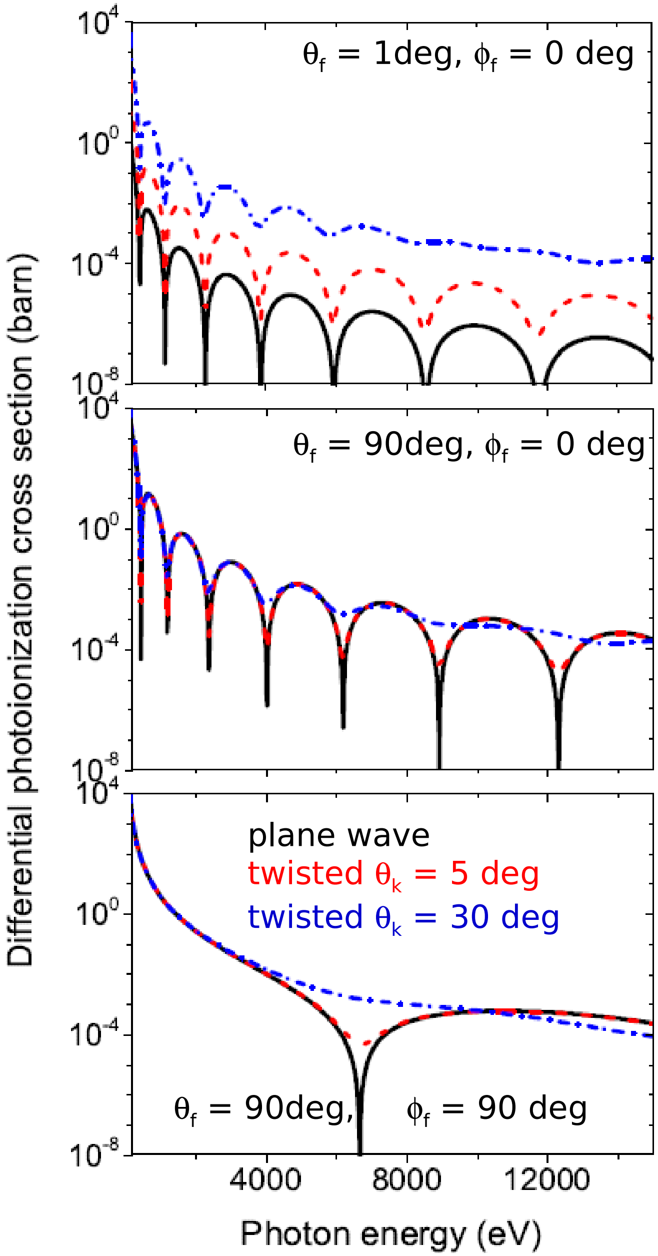
How does the cross sections differ for twisted light ?

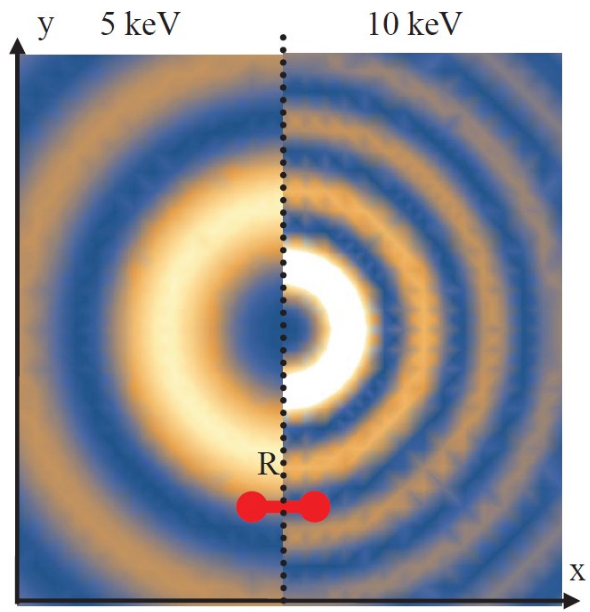
$$\boldsymbol{A}(\boldsymbol{r}) = \int a_{\kappa m}(\boldsymbol{k}_\perp)\, u_{k\lambda}\, e^{ikr}\, \frac{d^2\boldsymbol{k}_\perp}{(2\pi)^2}$$

$$M_{if} = -\frac{i}{c}\int \Psi_f^\dagger(\boldsymbol{r})\, \boldsymbol{\alpha}\, \boldsymbol{A}(\boldsymbol{r})\Psi_i(\boldsymbol{r})d\boldsymbol{r}$$

Photoionization of diatomic molecules

– ionization by twisted light

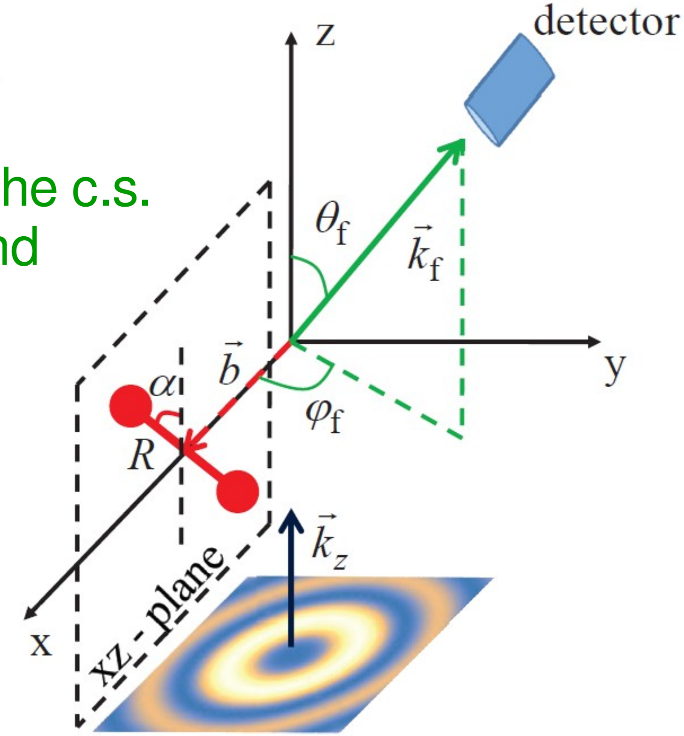




Intensity pofile for hν = 5 keV and 10 keV photons.

Internuclear distance R = 1.32 A

No oscillations remains in the c.s. for large opening angles and at higher photon energies.



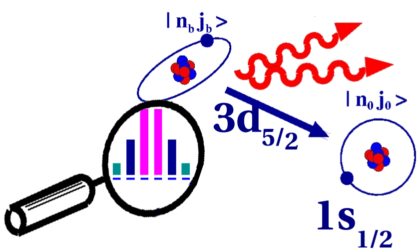
Ionization of H_2^+ if aligned under an angle 45° w.r.t. the z-axis (quantization axis).

A. Peschkov *et al.*, PRA 92 (2015) 043415.

Recent research activities of the group

i) Structure and properties of atoms & HCI

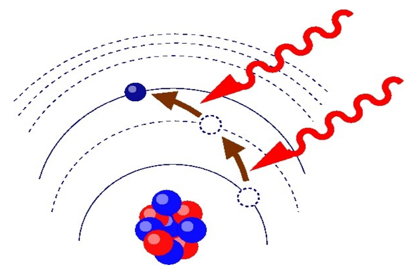
-- capture, excitation and ionization processes in strong static fields; accurate atomic calculations

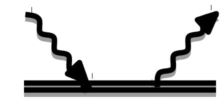


PRA 93 (2016) 063413.
PRC 93 (2016) 064318.
NJP 18 (2016) 103034.

iii) Photoionization and photon scattering

-- direct and sequential (multiple) ionization, Rayleigh scattering, polarization transfer

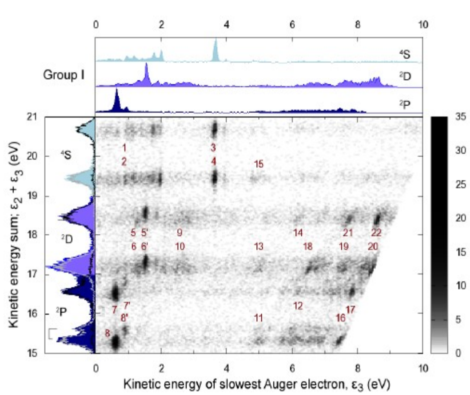




PRA 93 (2016) 023418.
PRA 93 (2016) 033409.
PRA 94 (2016) 032503.

ii) Auger cascades of light & medium elements

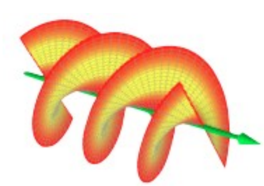
– systematic treatment of shake-processes; multiple photo-detachment, electrons in plasma



PRE 93 (2016) 061201(R).
PRA 94 (2016) 041401(R).

vi) Interactions with twisted light & electrons

-- photo-excitation, ionization, Compton scattering, Mott scattering by Bessel & Laguerre Gaussian beams



EPL 115 (2016) 410010.
PRA 94 (2016) 033420.
PRA 94 (2016) 041402(R).

Kassel -- Darmstadt -- Heidelberg – Oulu – Jena

Summary: combination of quite different (many-particle) techniques

- Schrödinger equation
- Dirac equation
- Quantum electrodynamics
- Group theory
- Density matrix theory

→ Study of atomic processes and quantum dynamics

Combination of different concepts

Use of systematic approximations to describe the behaviour of quantum systems:

- Density matrix techniques and spherical tensors
- Racah's algebra
- Multiconfigurational expansions (CI, MCDF)
- Many-body perturbation theory (MBPT, CC)
- Green's functions