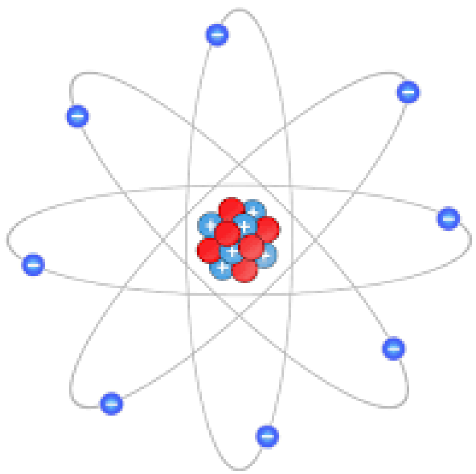
Atoms in strong and twisted fields



Stephan Fritzsche
Helmholtz-Institut Jena &
Theoretisch-Physikalisches Institut Jena
11th June 2019

seit 1558

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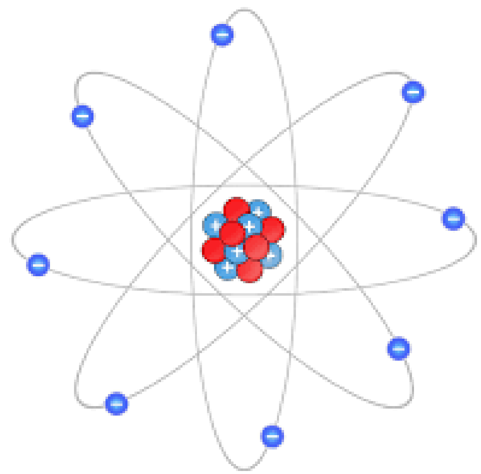
Helmholtz Institute Jena

Atoms in strong and twisted fields



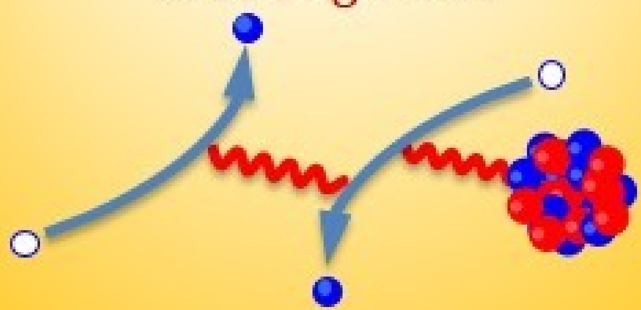
Why atoms ?

- Fundamental for „quantum mechanics“.
- Well-known „atomic interactions“: QED + atomic shell model.
- Precision spectroscopy & experiments.
- Variety of processes: Atoms are simple to manipulate & control.
- Help & support for many research areas.

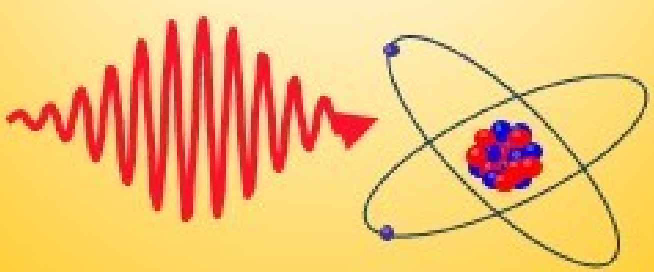
Atomic physics is still a „great playground“ for new ideas & concepts !

Atoms in strong and twisted fields

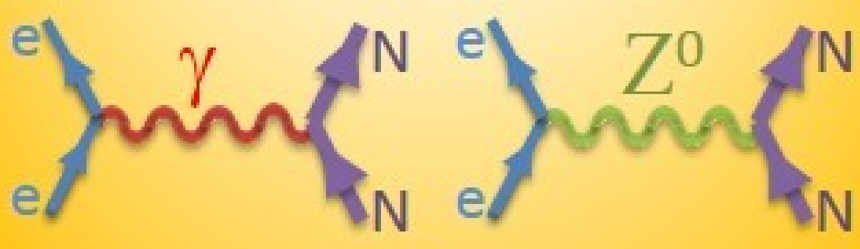
Correlated many-electron dynamics in strong fields



Relativistic photon-matter interaction in intense laser fields



Atomic structure of atoms and ions & fundamental symmetries

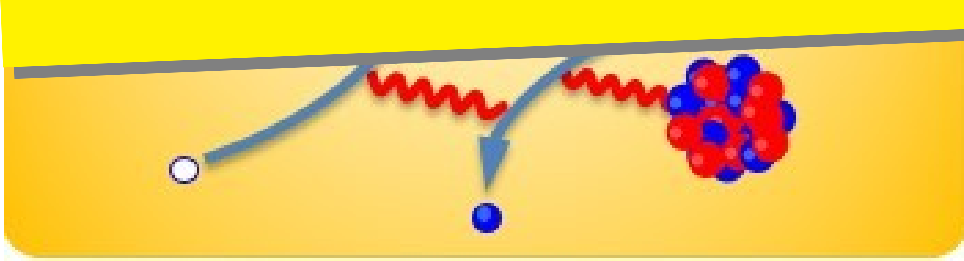


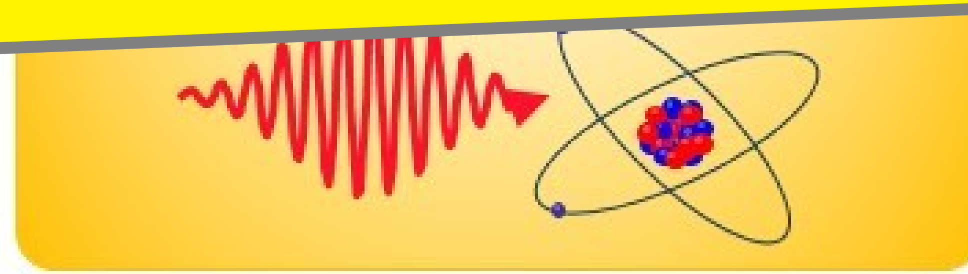
Supercritical fields in ultra-relativistic or slow ion-ion collisions

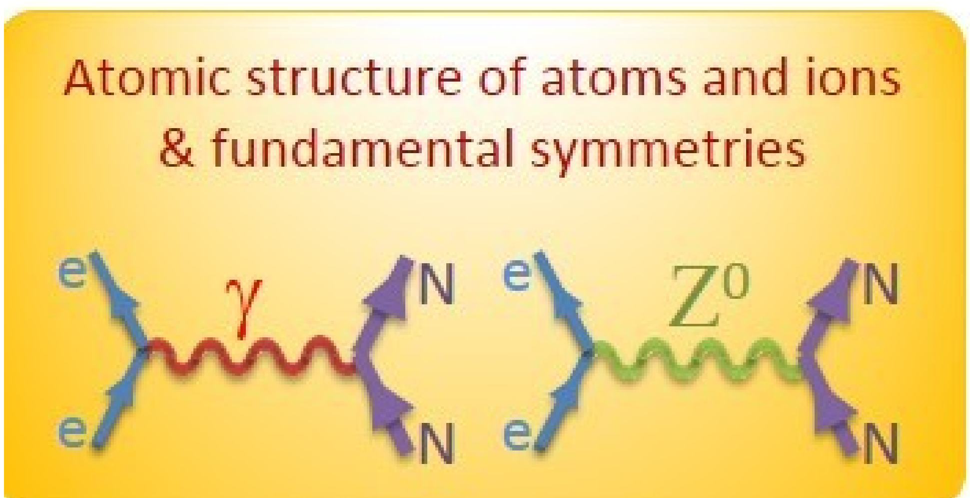
Atoms in strong and twisted fields

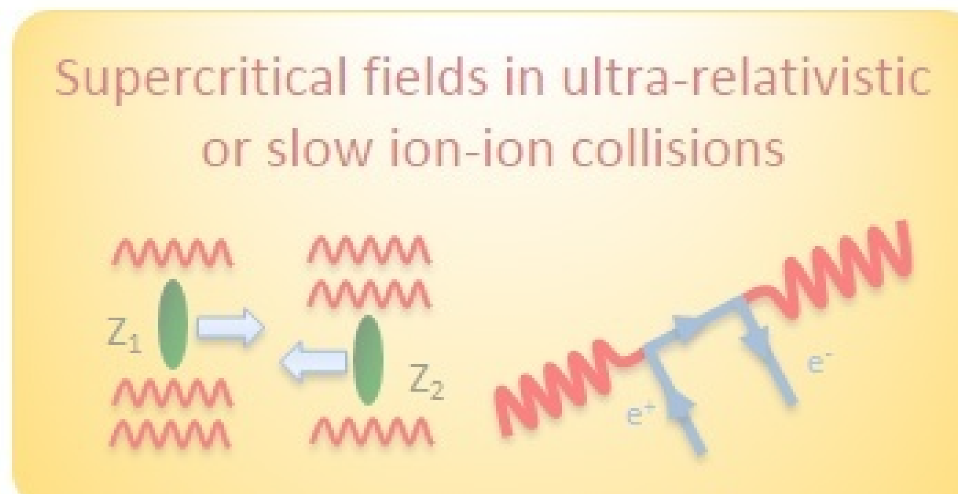
Question:

How well can be describe and (theoretically) predict the structure and behaviour of atoms in different environments ?

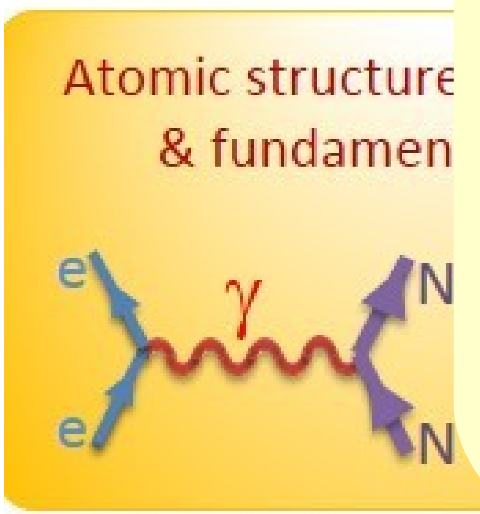








Atoms in strong and twisted fields



Question:

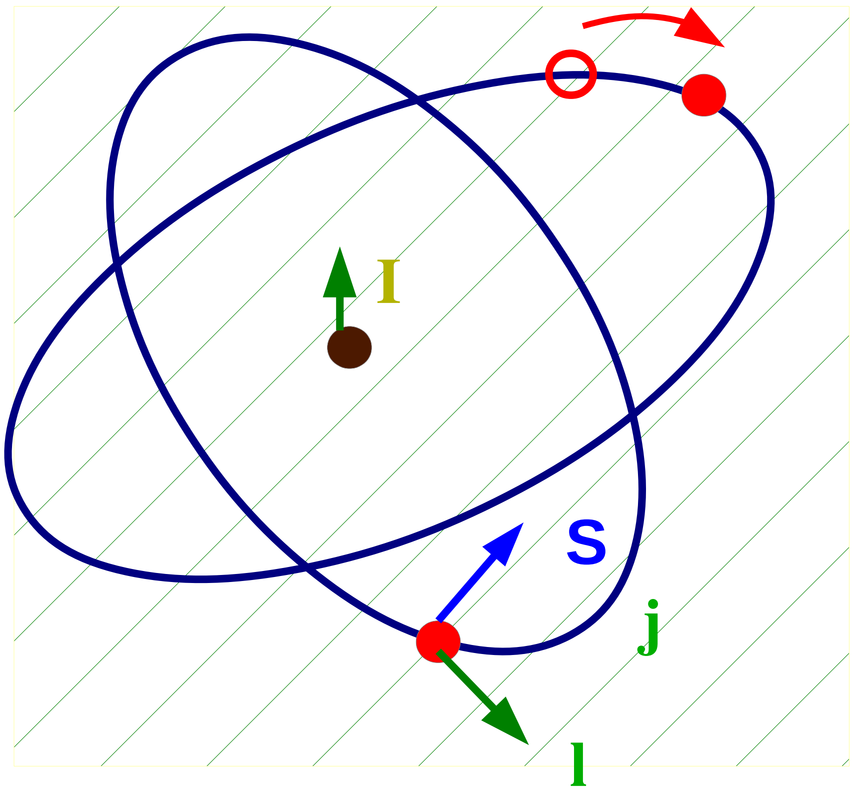
How well can be describe and (theoretically) predict the structure and behaviour of atoms in different environments ?

Plan of this talk

- Atomic physics @ GSI: A (very) short reminder
- Quiz: Atomic processes in a nutshell
- Demands from experiment & theory
- Jena Atomic Calculator (JAC): A fresh approach ...
- Atoms in twisted beams: Ionization & HHG
- Summary & conclusions

Atomic interactions are (beliefed to be) known

– although not easy to get under good control



External fields

Nuclear potential

Instantaneous Coulomb repulsion
between all pairs of electrons

Spin-orbit interaction

Relativistic electron velocities;
magnetic contributions and
retardation

QED: radiative corrections

Hyperfine structure

Electric and magnetic nuclear
moments (isotopes)

Motion of the nucleus: Reduced
mass and mass polarization

Atomic interactions are (beliefed to be) known

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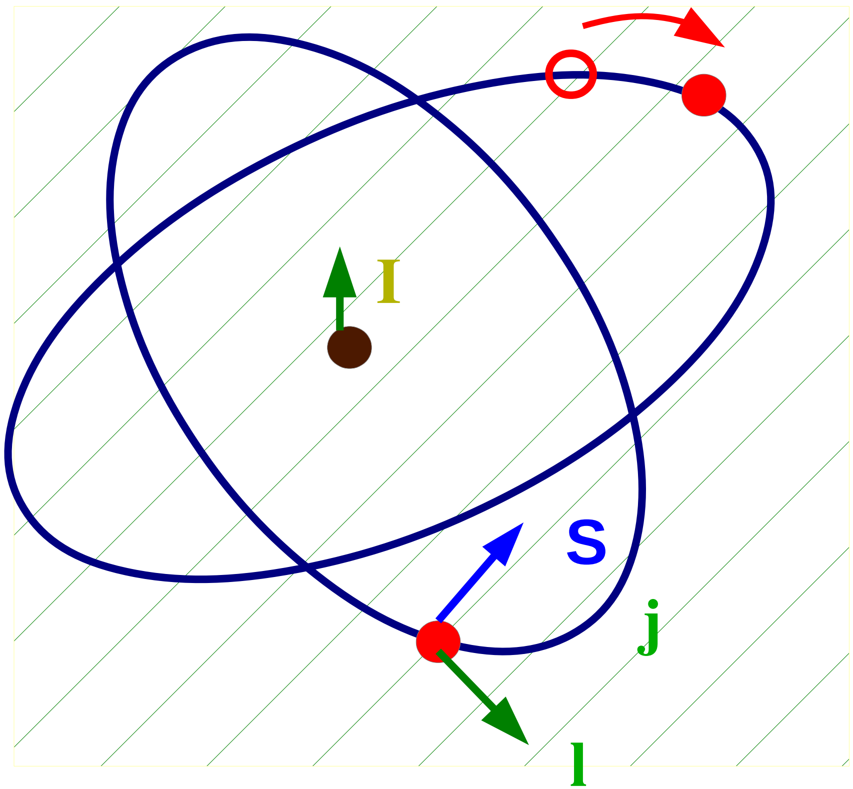

External fields

Nuclear potential

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QED: radiative corrections

Hyperfine structure

Electric and mag[...]uclear moments (iso[...]s)

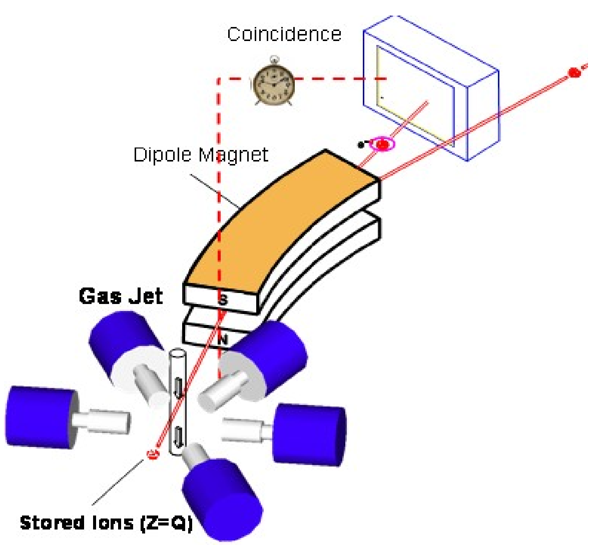
Perturbation theory

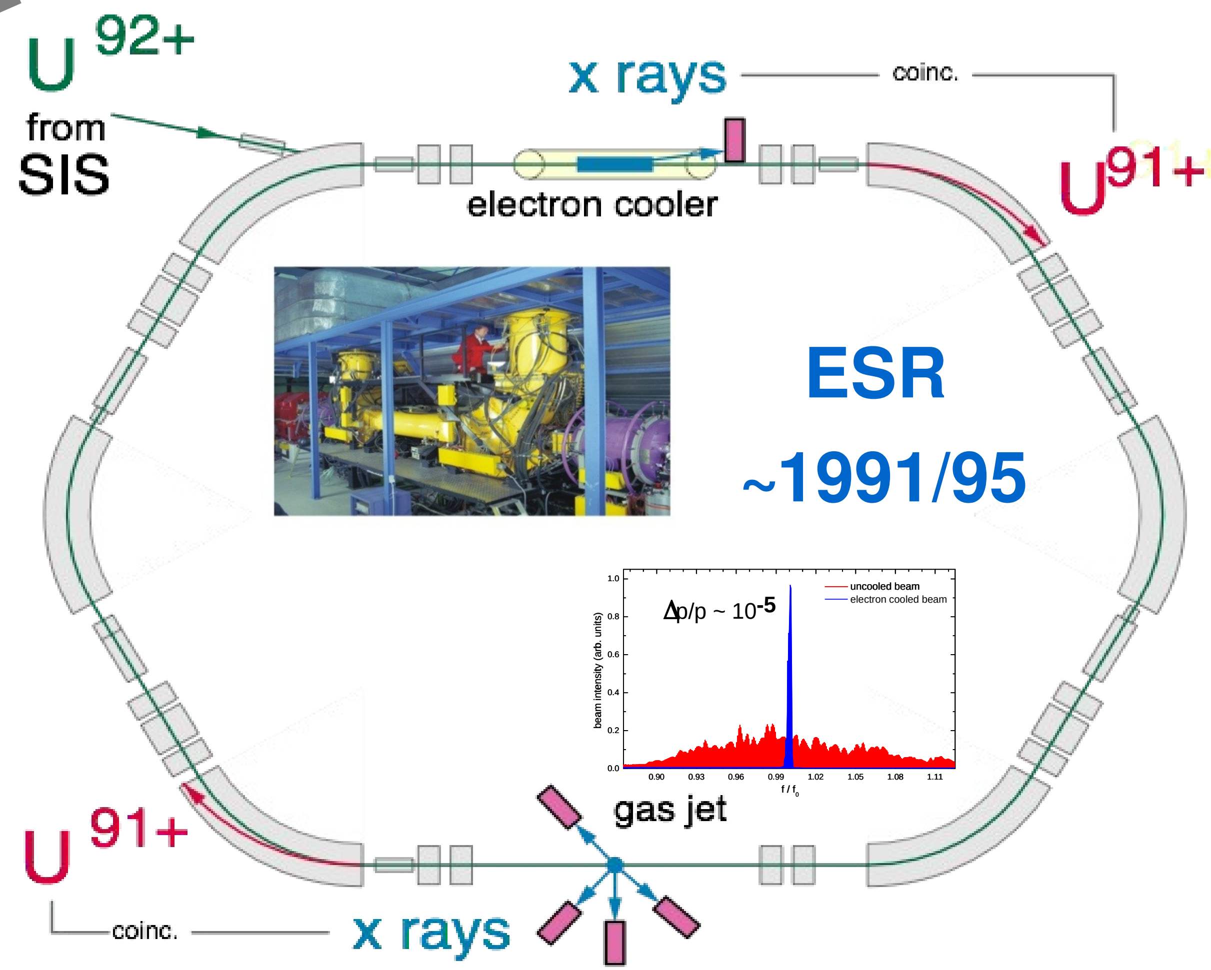
Motion of the nucleus: Reduced mass and mass polarization

Interaction with radiation & external fields; collisions.

Atomic excitations in relativistic heavy-ion collisions

Successful experiments ~~for the last~~ 20 years ago !

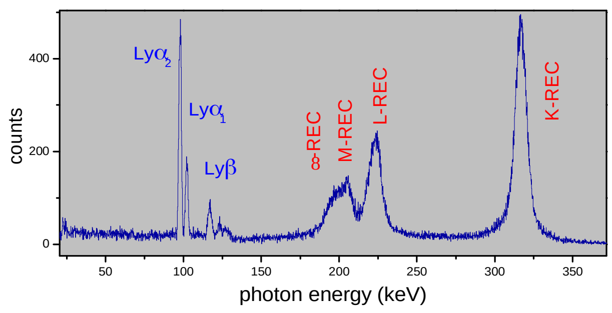


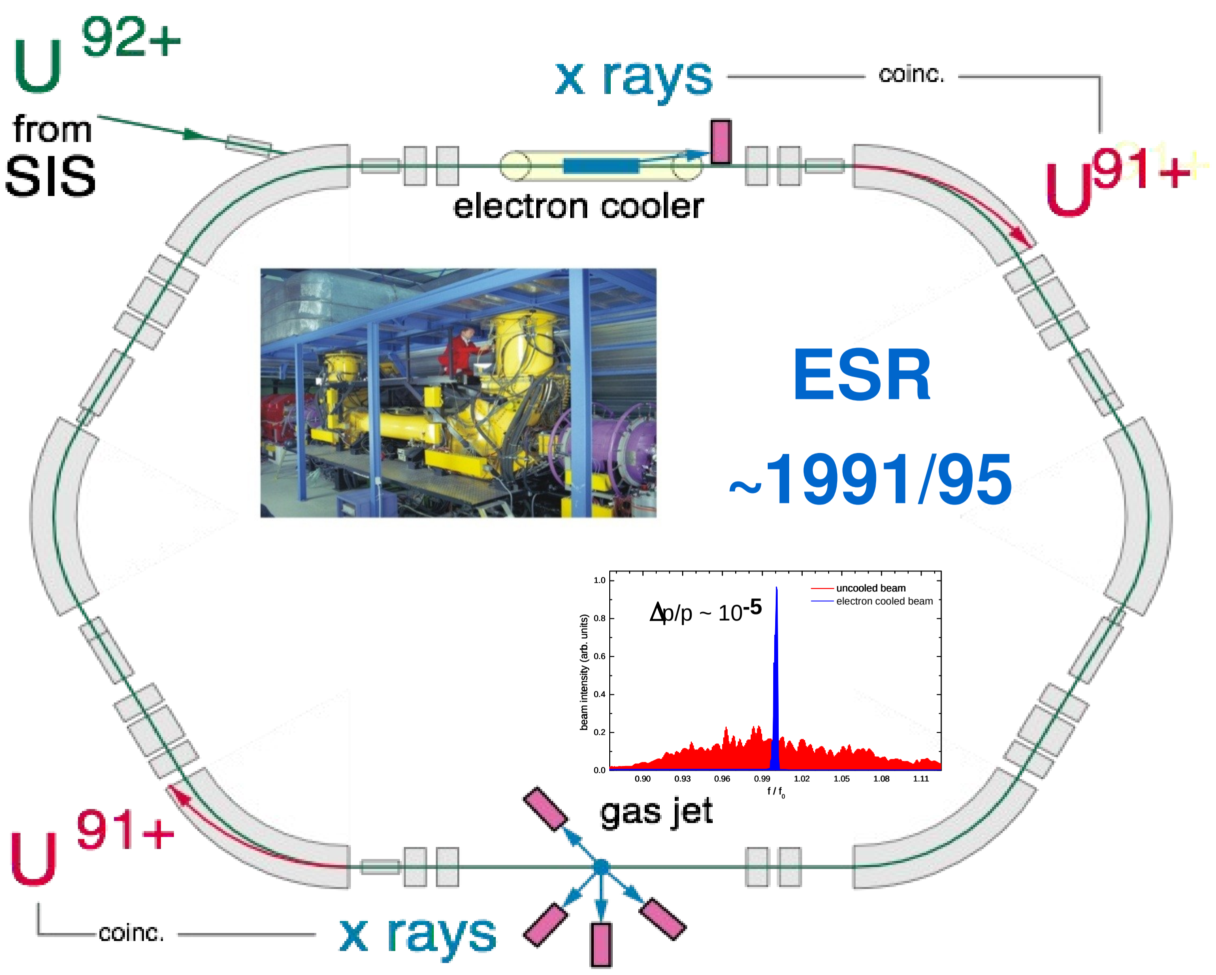


J. Eichler and T. Stöhlker, Phys. Reports 439 (2007) .

Atomic excitations in relativistic heavy-ion collisions

$U^{92+} + N_2$ @ 295 MeV/u





X-ray emission due to:

- Radiative electron capture (RR & REC)
- Characteristic transitions (Ly-α & K-α)
- Dielectronic recombination
- Coulomb excitation & ionization
- ...

J. Eichler and T. Stöhlker, Phys. Reports 439 (2007) .

Atomic excitations in relativistic heavy-ion collisions

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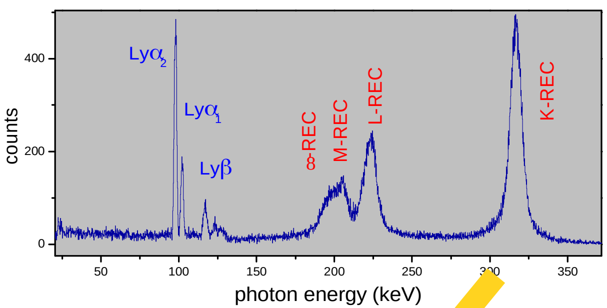
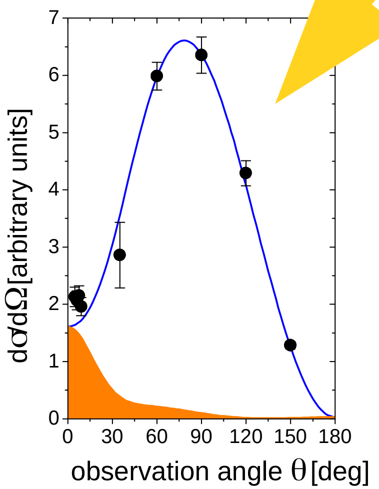
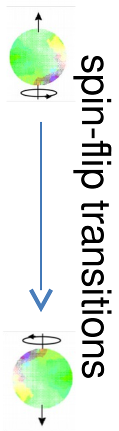
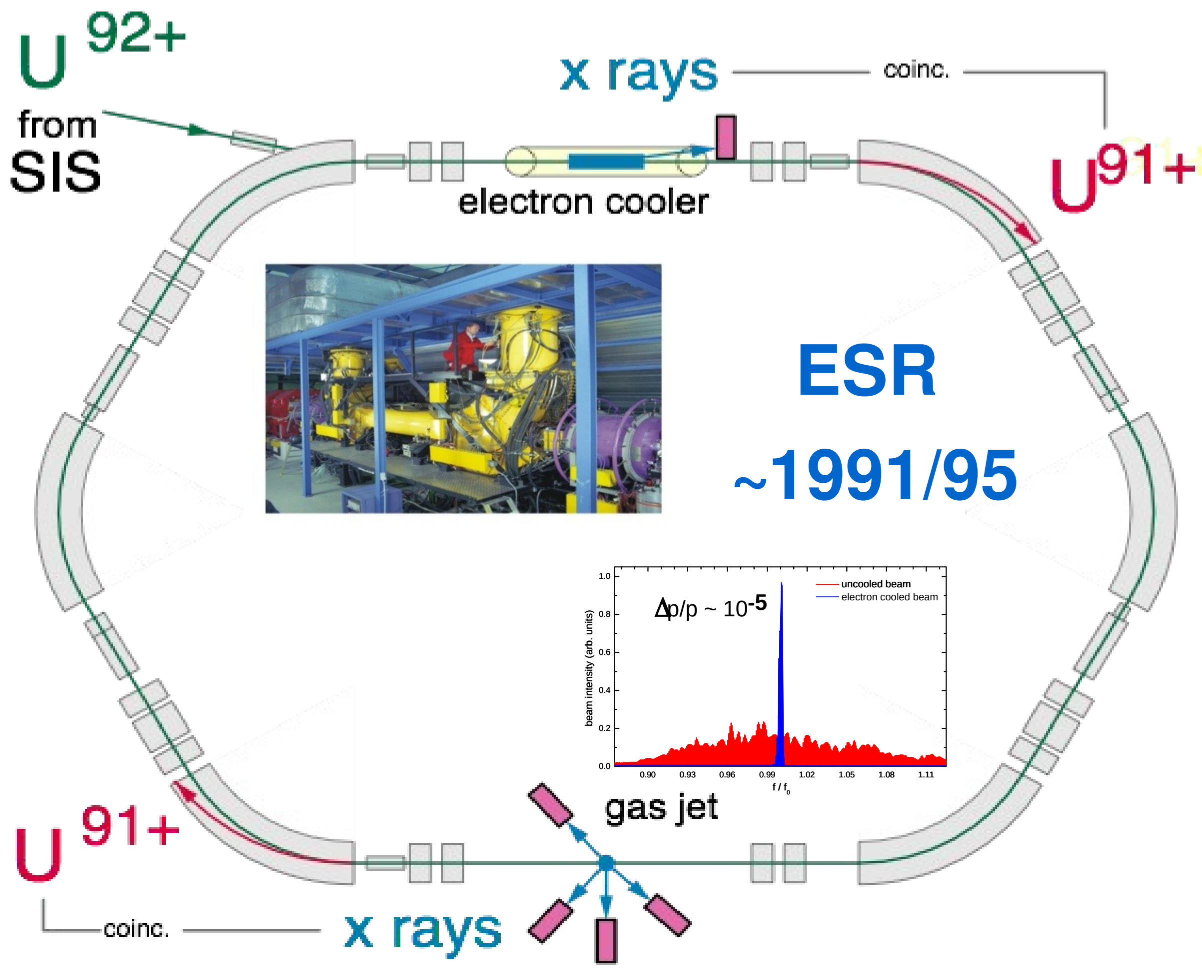


Photon angular distribution for REC into the K-shelll (U^{92+}, 310 MeV/u)







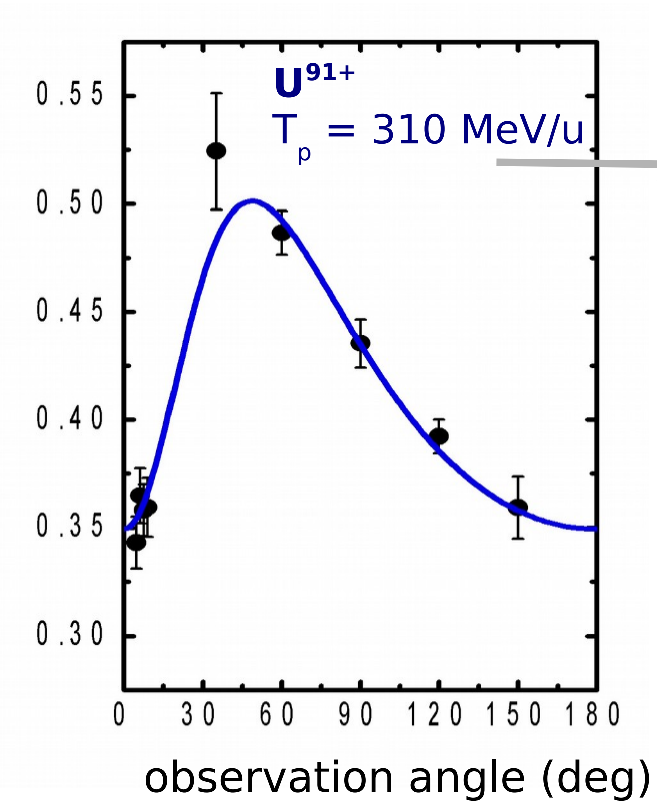
Identification of spin-flip transitions.

T. Stöhlker et al., PRL 79 (1997) 3270.
J. Eichler and T. Stöhlker, Phys. Reports 439 (2007) .

A. Surzhykov et al. PRL 88 (2002) 153001.

Atomic excitations in relativistic heavy-ion collisions

1 Alignment of the $2p_{3/2}$ state



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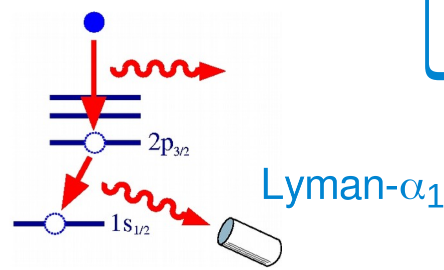
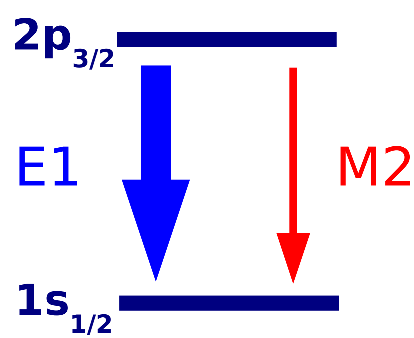
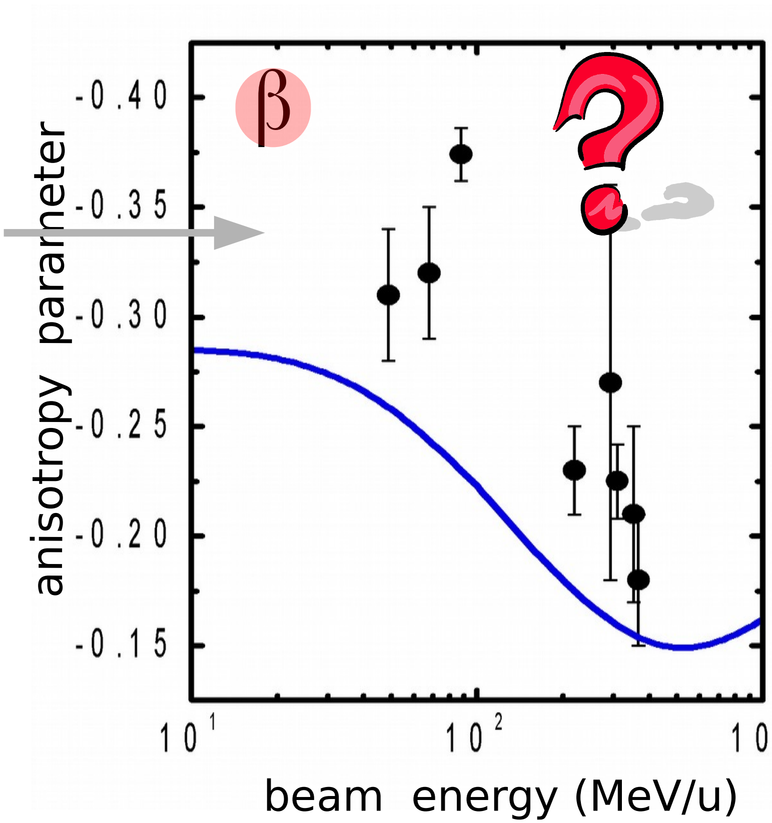


$$W(\theta) \propto 1 + \beta P_2(\cos\theta)$$



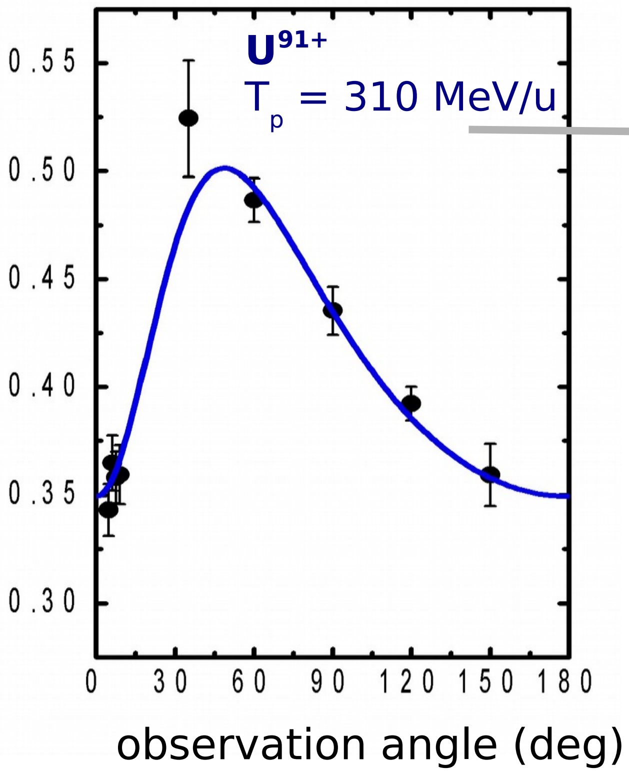


J. Eichler et al., PRA 58 (1998) 2128.

- Fundamental (relativistic) interactions in strong Coulomb field ?
- Virtual vs. real photon fields ?

A. Surzhykov et al. PRL 88 (2002) 153001.
S. Fritzsche et al. PRL 103 (2009) 113001.

Atomic excitations in relativistic heavy-ion collisions



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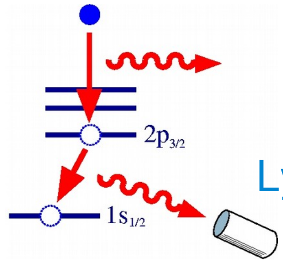

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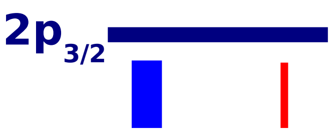

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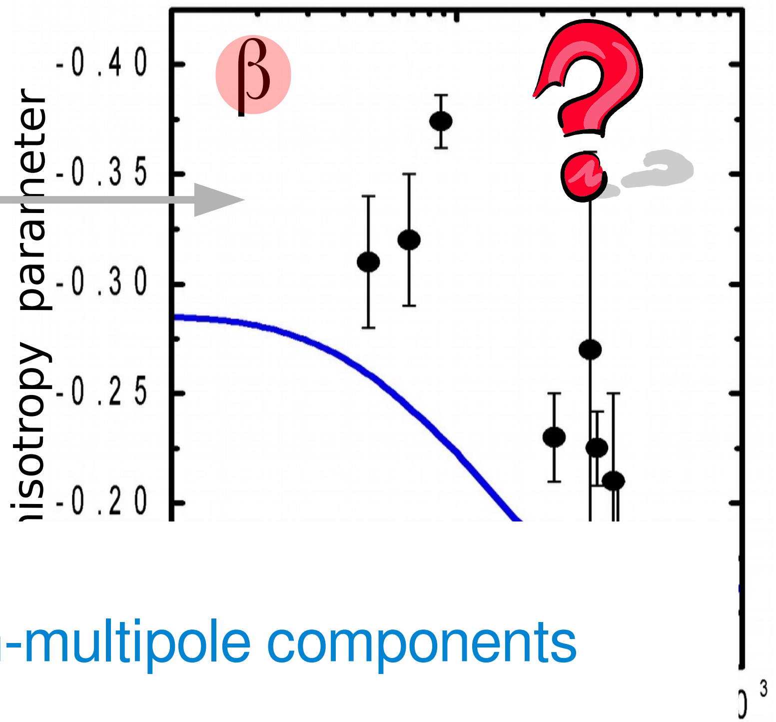


2 Probe for high-multipole components

3 Can one „measure“ multipole fields ?

4 Lyman-α vs. K-α emission

5 K-α emission after Coulomb excitations

...

- Fundament
- Virtual vs. re

Quiz: Atomic processes in a nutshell

-- for “intermediates” in atomic and plasma physics

$$A + n\,\hbar\omega \longrightarrow A^{+(*)} + e_p^-$$

$$A + n\,\hbar\omega \longrightarrow A^{+(*)} + (e_{p_1}^- + e_{p_2}^-)$$

$$A^{q+} + e_s^- \longrightarrow A^{(q-1)+} + \hbar\omega$$ ... radiative recombination

$$A^{q+} + e_s^- \longrightarrow A^{(q-1)+\,*} \longrightarrow A^{(q-1)+\,(*)} + \hbar\omega$$ ... dielectronic recombination

$$A + \hbar\omega \longrightarrow A^{(*)} + \hbar\omega'$$

$$A^{q+*} \longrightarrow A^{(q+1)+(*)} + (e_a^- + \hbar\omega)$$

$$A^{q+*} \longrightarrow A^{(q+2)+(*)} + (e_{a_1}^- + e_{a_2}^-)$$

$$A + \hbar\omega \longrightarrow A^* \longrightarrow A^{(*)} + \hbar\omega'$$

$$A + \hbar\omega \longrightarrow A^{+,*} + e_p^- \longrightarrow A^{(*)} + e_p^- + \hbar\omega'$$

$$A + Z_p \longrightarrow A^* + Z_p'$$

$$A^{(q+1)+} + Z_p \longrightarrow A^{(q+1)+\,(*)} + e^- + Z_p'$$

Quiz: Atomic processes in a nutshell

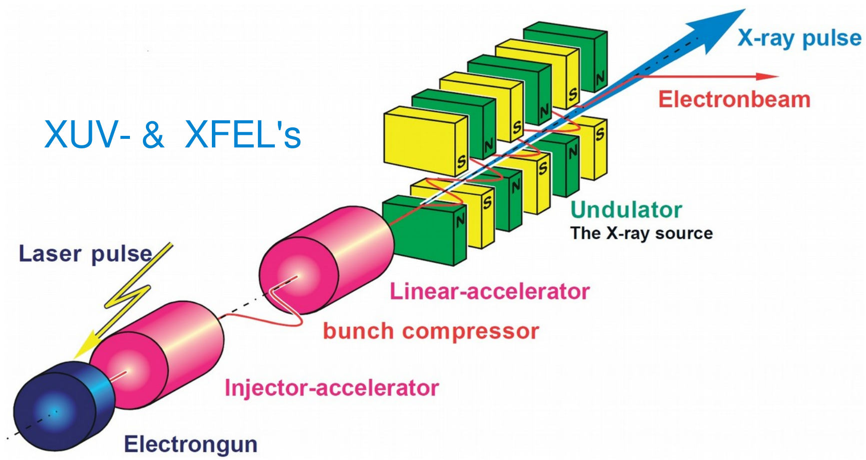
-- for "intermediates" in atomic and plasma physics

$A + n\,\hbar\omega$	$\longrightarrow \quad A^{+(*)} + e_p^-$	... multi − photon ionization
$A + n\,\hbar\omega$	$\longrightarrow \quad A^{+(*)} + (e_{p_1}^- + e_{p_2}^-)$	... multi − photon double ionization
$A^{q+} + e_s^-$	$\longrightarrow \quad A^{(q-1)+} + \hbar\omega$	... radiative recombination
$A^{q+} + e_s^-$	$\longrightarrow \quad A^{(q-1)+\,*} \longrightarrow A^{(q-1)+\,(*)} + \hbar\omega$	... dielectronic recombination
$A + \hbar\omega$	$\longrightarrow \quad A^{(*)} + \hbar\omega'$	... Rayleigh/Compton
A^{q+*}	$\longrightarrow \quad A^{(q+1)+(*)} + (e_a^- + \hbar\omega)$	... radiative Auger
A^{q+*}	$\longrightarrow \quad A^{(q+2)+(*)} + (e_{a_1}^- + e_{a_2}^-)$	... double Auger
$A + \hbar\omega$	$\longrightarrow \quad A^* \longrightarrow A^{(*)} + \hbar\omega'$	... photo − excitation & fluorescence
$A + \hbar\omega$	$\longrightarrow \quad A^{+,*} + e_p^- \longrightarrow A^{(*)} + e_p^- + \hbar\omega'$	... photo − ionization & fluorescence
$A + Z_p$	$\longrightarrow \quad A^* + Z_p'$	... Coulomb excitation
$A^{(q+1)+} + Z_p$	$\longrightarrow \quad A^{(q+1)+\,(*)} + e^- + Z_p'$	... Coulomb ionization

- Indeed, these and many other processes occur in atomic, plasma and astro physics as well as at various places elsewhere.
- How much help can atomic theory provide ? -- Which tools are available ?

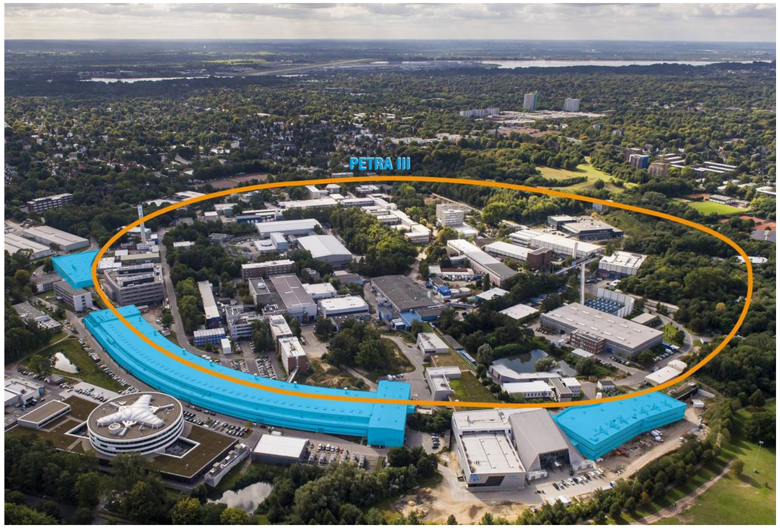
Demands from experiment

– owing to new large-scale facilities



space missions

synchrotrons



Demands from experiment

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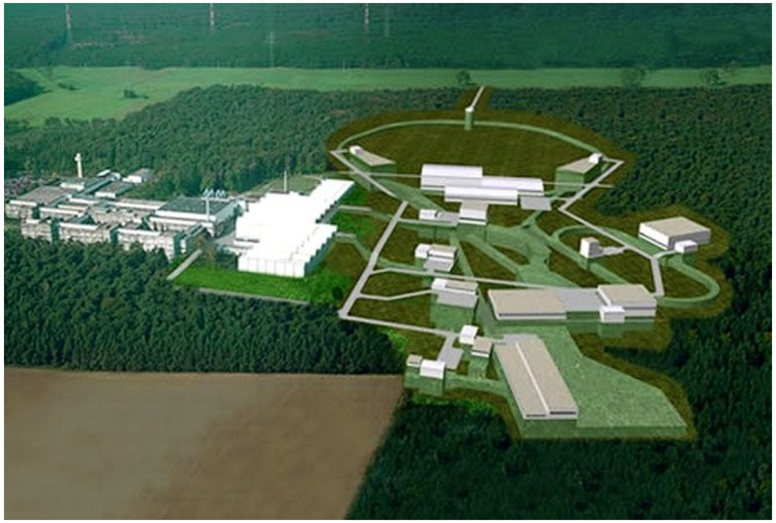
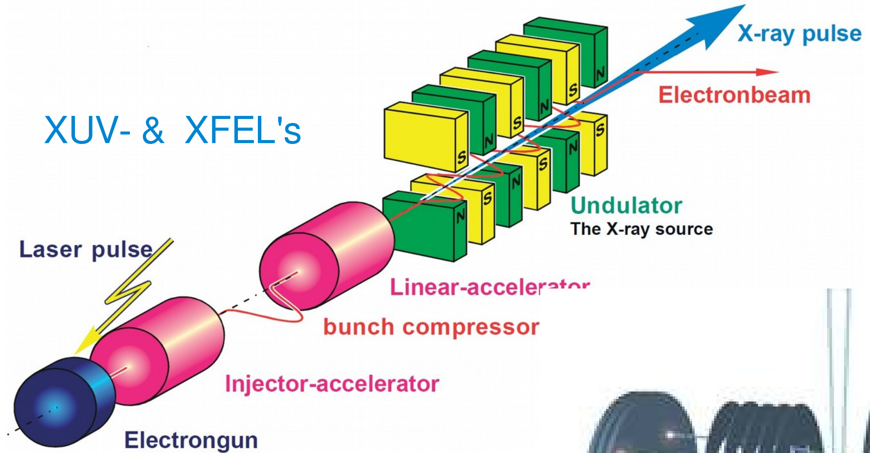
space missions

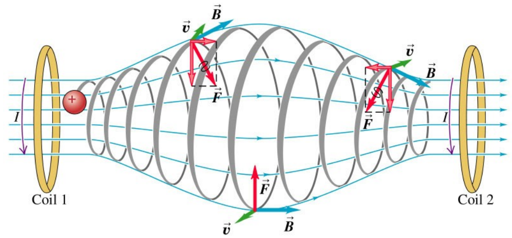


Demands from experiment

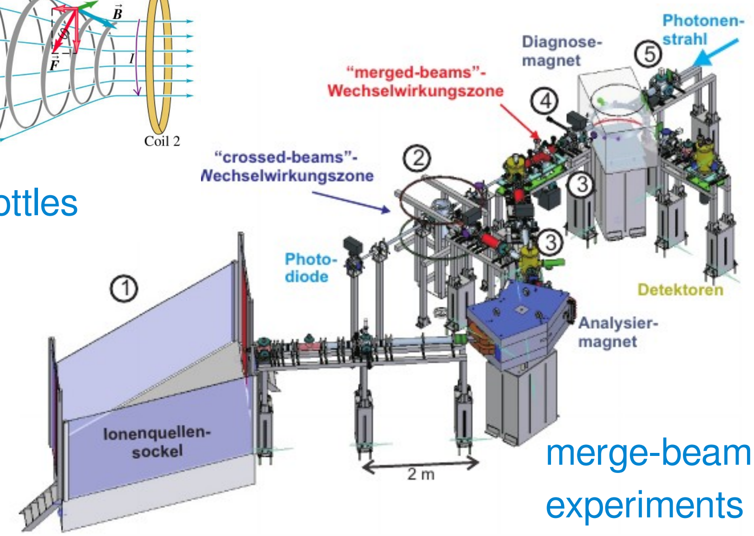
– owing to new large-scale facilities & detector developments

XUV- & XFEL's

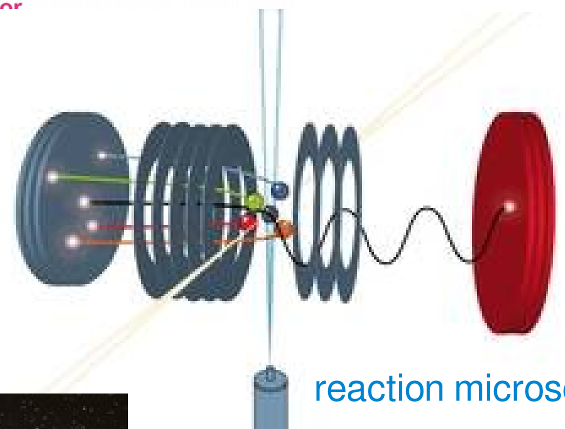




magnetic bottles

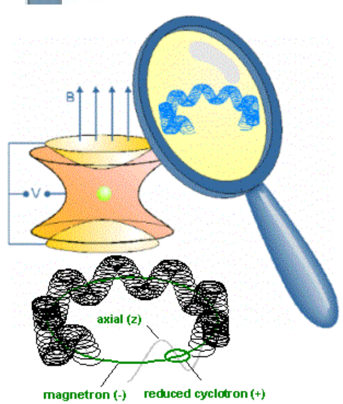


merge-beam experiments



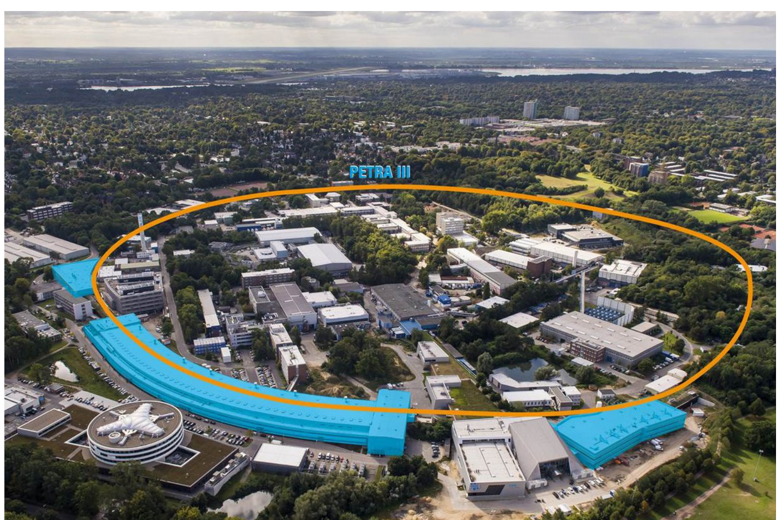
reaction microscopes

space missions



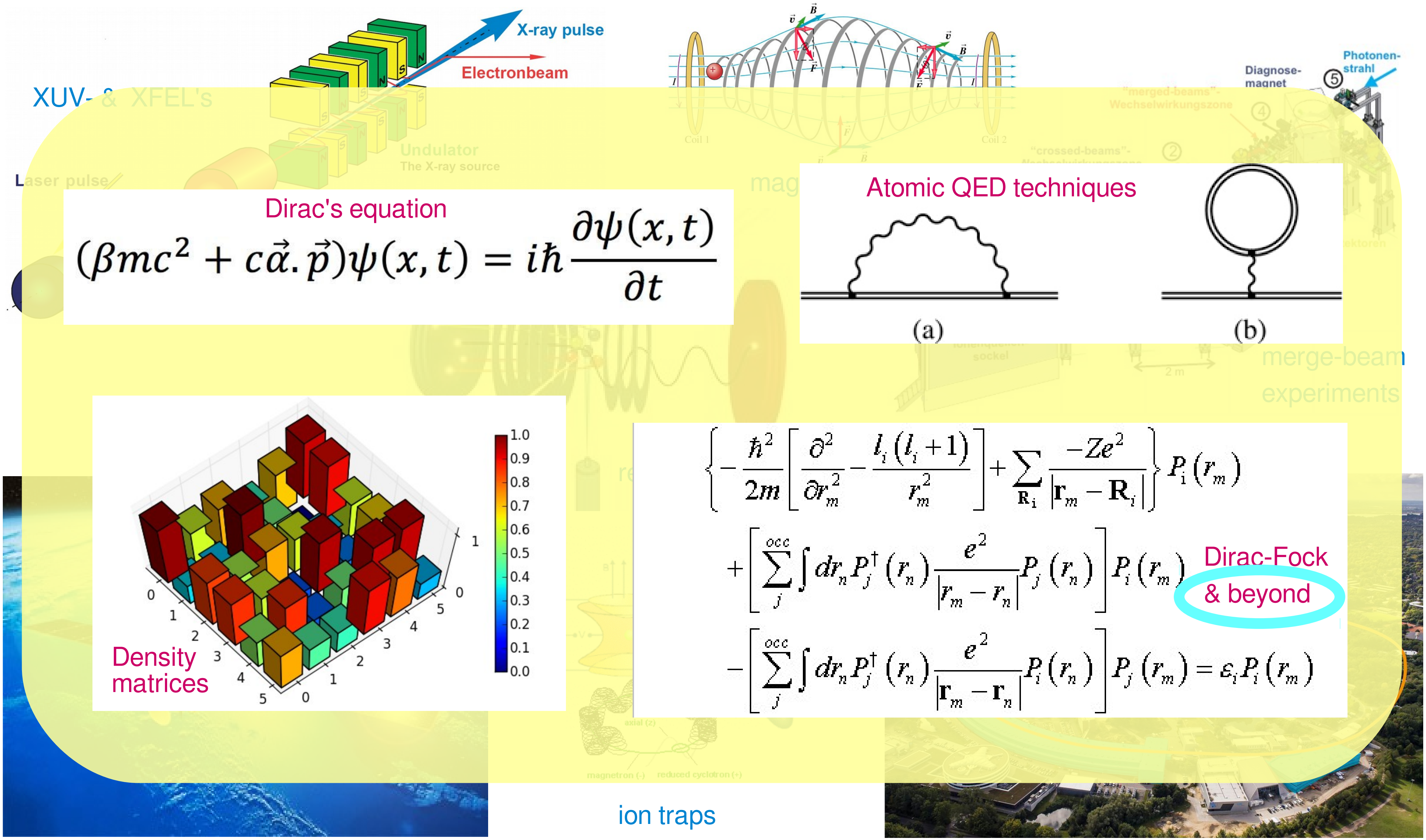
ion traps

synchrotrons



Demands from experiment

– owing to new large-scale facilities & detector developments



Dirac's equation

$$(\beta mc^2 + c\vec{\alpha}.\vec{p})\psi(x,t) = i\hbar\frac{\partial\psi(x,t)}{\partial t}$$

$$\left\{-\frac{\hbar^2}{2m}\left[\frac{\partial^2}{\partial r_m^2} - \frac{l_i(l_i+1)}{r_m^2}\right] + \sum_{\mathbf{R}_i}\frac{-Ze^2}{|\mathbf{r}_m - \mathbf{R}_i|}\right\}P_i(r_m) + \left[\sum_j^{occ}\int dr_n P_j^\dagger(r_n)\frac{e^2}{|r_m - r_n|}P_j(r_n)\right]P_i(r_m) - \left[\sum_j^{occ}\int dr_n P_j^\dagger(r_n)\frac{e^2}{|\mathbf{r}_m - \mathbf{r}_n|}P_i(r_n)\right]P_j(r_m) = \varepsilon_i P_i(r_m)$$

Demands from experiment

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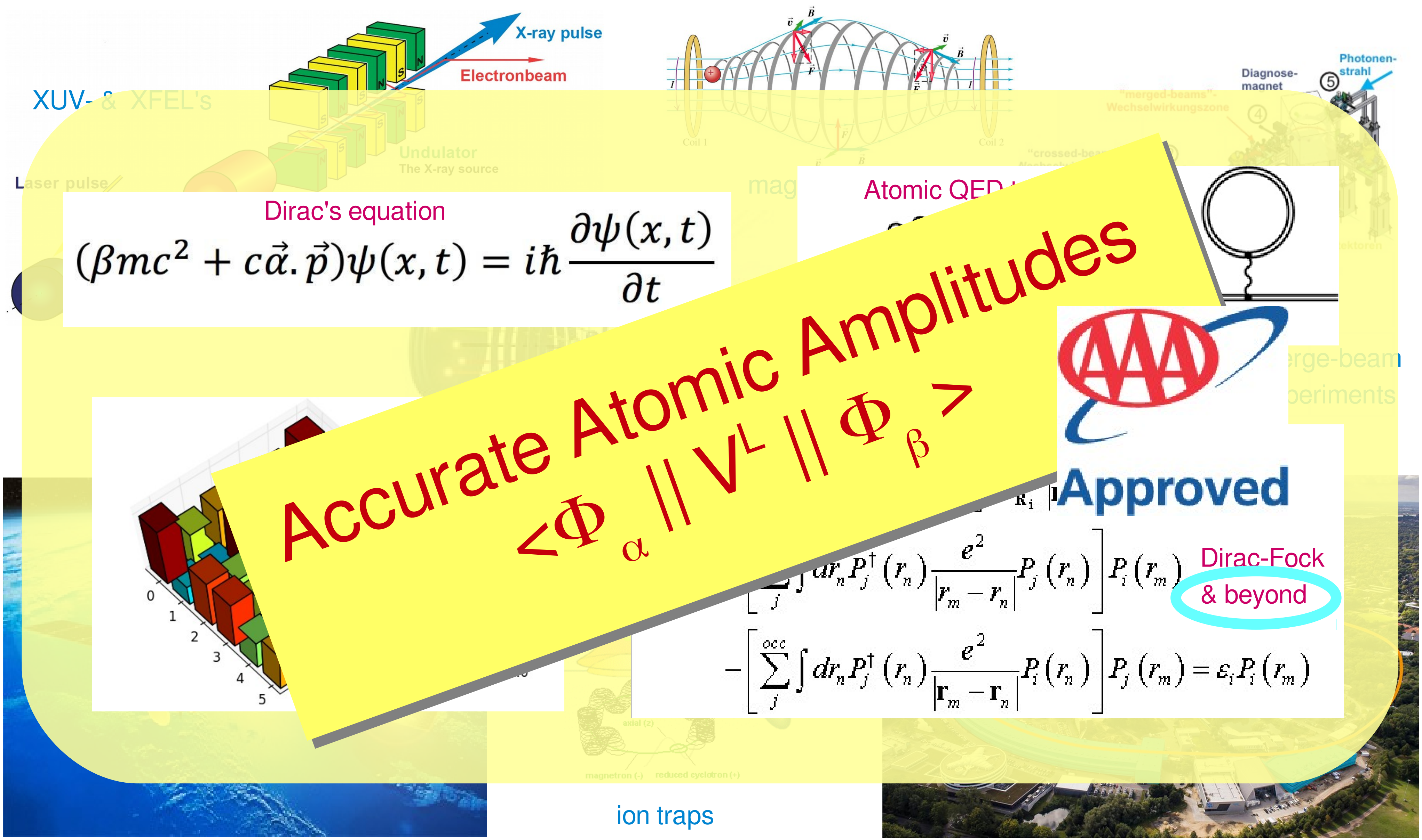

https://www.github.com/OpenJAC/JAC.jl

Jena Atomic Calculator (JAC)

JAC ... Jena atomic calculator provides tools for performing atomic (structure) calculations at various degrees of complexity and sophistication. ... JAC also facilitates interactive computations, the simulation of atomic cascades, the time-evolution of statistical tensors as well as various semi-empirical estimates of atomic properties. In addition, the Jac module supports the graphical representation of level energies, electron and photon spectra, radial orbitals and others.

Central questions to any new implementation:

- Is a common (and community) platform for atomic computations desirable ?
- How can we benefit from a good 'core machinery' ?
- How simple and user-friendly can it be made ?
- How to combine productivity & performance in developing such a platform ?

Atomic cascades

- Average singe-configuration approach
- Multiple-configuration approach
- Incorporation of shake-up & shake-off
- Ion & electron distributions, ...

Open-source applications in physics, science and technology.

Semi-empirical estimate

- Weak-field ionization rates
- Stopping powers
- Plasma Stark broadening, ...

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Atomic properties

- Hyperfine splitting & representation
- Zeeman splitting; Lande factors
- Isotope shifts, atomic for factors
- Plasma shifts, α-variations
- Approximate Greens function, ...

Atomic processes

- Photon emission & transition probabilities
- Photoexcitation, ionization & recombinat.
- Auger emission & di-electr. recombination
- Rayleigh-Compton scattering
- Multiphoton (de-) excitation, ...

Atomic cascades

- Average singe-configuration approach
- Multiple-configuration approach
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- Ion & electron distributions, ...

Interactive High-Level Language

JAC

Jena Atomic Calculator

A Julia implementation for atomic computations.

Open-source applications in physics, science and technology.

Atomic responses

- Field-induced processes & ionization
- High-harmonic generation
- Particle-impact processes

Atomic time-evolution

- Liouville equation for statistical tensors & atomic density matrices
- Atoms in intense light pulses
- Angle- & polarization-dependent observables

Semi-empirical estimate

- Weak-field ionization rates
- Stopping powers
- Plasma Stark broadening, ...

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Atomic cascades

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- Ion & electron distributions, ...

Atomic responses

Why Julia ?

- (Very) fast, high-level language (from MIT, since ~ 2012).
- Combines productivity and performance.
- Multiple dispatch ... to distinguish generic code, still dynamic.
- Just in-time (JIT) compilation, fast loops.
- Rapid code development: no linkage; in-built benchmarking.
- Most code & macros are written in Julia.
- Extensive list of packages.
- No storage management, little declaration; type stability.
- Easy documentation, ...

https://www.github.com/OpenJAC/JAC.jl

README.md

build error | build passing | codecov unknown | coverage unknown

Jena Atomic Calculator (JAC) for the computation of atomic structures, processes and cascades

What is JAC?

We here provide a first public version of *JAC*, the Jena Atomic Calculator and an open-source Julia package for doing atomic computations. JAC is a (relativistic) electronic structure code for the computation of (many-electron) interaction amplitudes, properties as well as a large number of excitation and decay processes for open-shell atoms and ions across the whole periodic table. In the future, moreover, JAC will -- more and more -- facilitate also studies on atomic cascades, responses as well as the time-evolution of atoms and ions.

A primary guiding philosophy of JAC was to develop a **general and easy-to-use toolbox for the atomic physics community**, including an interface that is equally accessible for working spectroscopiest, theoreticians and code developers. Beside of its simple use, however, I also wish to provide a modern code design, a reasonable detailed documentation of the code and features for integrated testing. In particular, most typical calculations and the handling of atomic data should appear within the code similar to how they would appear in spoken or written language. Shortly speaking, JAC aims to provide a powerful **platform for daily use and to extent atomic theory towards new applications**.

Kinds of computations

In some more detail, JAC distinguishes and aims to support (partly still in the future) **seven kinds of computations** which can be summarized as follows:

1. **Atomic computations**, based on explicitly specified electron configurations: This kind refers to the computation of level energies, atomic state representations and to either one or several atomic properties for selected levels of a given multiplet. It also help compute *one* selected process at a time, if atomic levels from two or more multiplets are involved in atomic transitions.
2. **Restricted active-space computations (RAS)**: This kind concerns systematically-enlarged calculations of atomic states

Quickstart

The numerous features of JAC can be easily understood by following the tutorials that are distributed together with the code. Further details can then be found from the Manual, Compendium & Theoretical Background to JAC. Make use the index or a full-text search to find selected items in this (.pdf) manual.

A very first **simple example** has been discussed in the reference above and refers to the low-lying level structure and the Einstein A and B coefficients of the 3s 3p^6 + 3s^2 3p^4 3d → 3s^2 3p^5 transition array for Fe^9+ ions, also known as the spectrum Fe X. To perform such a computation within the framework of JAC, one needs to specify the initial- and final-state configurations in an instance of an `Atomic.Computation`, together with the specifier `process=RadiativeX`. We here also provide a title (line), the multipoles (default E1) and the gauge forms for the coupling of the radiation field that are to be applied in these calculations:

```
comp = Atomic.Computation("Energies and Einstein coefficients for the spectrum Fe X",  Nuclear.Model(26.);
                initialConfigs = [Configuration("[Ne] 3s 3p^6"), Configuration("[Ne] 3s^2 3p^4 3d")],
                finalConfigs   = [Configuration("[Ne] 3s^2 3p^5")],
                process = RadiativeX,
                processSettings = Radiative.Settings([E1, M1, E2, M2], [UseCoulomb, UseBabushkin] )
perform(comp::Atomic.Computation)
```

This example is discussed also in the tutorial.

Tutorials

The following IJulia/jupyter notebooks introduce the reader to JAC and demonstrate various features of this toolbox.
They can be explored statically at GitHub or can be run locally after the software repository has been cloned and installed. In order to modify the cell-output of the notebooks and to better print the *wide tables*, you can create or modify the file ~/.jupyter /custom/custom.css in your home directory and add the line: div.output_area pre { font-size: 7pt;} .

- Getting started
- Simple hydrogenic estimates
- Nuclear models and potentials
- Atomic potentials
- SCF + CI computations for carbon
- Einstein coefficients for Fe X

https://www.github.com/OpenJAC/JAC.jl

Jena Atomic Calculator (JAC)

-- A fresh approach to the computation of atoms, ...

JAC ... Jena atomic calculator provides tools for performing atomic (structure) calculations at various degrees of complexity and sophistication. ... JAC also facilitates interactive computations, the simulation of atomic cascades, the time-evolution of statistical tensors as well as various semi-empirical estimates of atomic properties. In addition, the Jac module supports the graphical representation of level energies, electron and photon spectra, radial orbitals and others.

Example: Einstein A and B coefficients for the Fe X spectrum;

$$Fe^{9+} \quad [Ne]\, 3s^2\, 3p^5 \rightarrow [Ne]\, 3s\, 3p^6 \; + \; 3s^2\, 3p^4\, 3d$$

```
> wa = Atomic.Computation("Fe X: Einstein",  NuclearModel(26.), ...,
            [Configuration("[Ne] 3s^2 p^5")], ...,
            [Configuration("[Ne ] 3s 3p^6"), Configuration("[Ne] 3s^2 3p^4 3d") ], ...,
            Radiative, Radiative.Settings([E1, M2], [UseCoulomb, UseBabushkin], false, false, ...  )
> perform(wa)

  ... in perform('computation: SCF', ...)
  Compute CI matrix of dimension 1 x 1 for the symmetry 1/2^+ ...   done.
  Compute CI matrix of dimension 1 x 1 for the symmetry 3/2^+ ...   done.
  ...
```

GUI ?
(graphical user interface)

https://www.github.com/OpenJAC/JAC.jl

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Example: Einstein A and B coefficients for the Fe X spectrum;

Fe^{9+} [Ne] $3s^2\, 3p^5 \rightarrow$ [Ne] $3s\, 3p^6 + 3s^2\, 3p^4\, 3d$

- Generation of start orbitals
- Computation of angular coefficients (on fly)
- Self-Consistent-Field (SCF) iteration
- Set-up and diagonalization of Hamiltonian matrix
- Breit, QED, many-body corrections, ...
- Compute all (many-electron) transition amplitudes

https://www.github.com/OpenJAC/JAC.jl

Jena Atomic Calculator (JAC)

-- A fresh approach to the computation of atoms, ...

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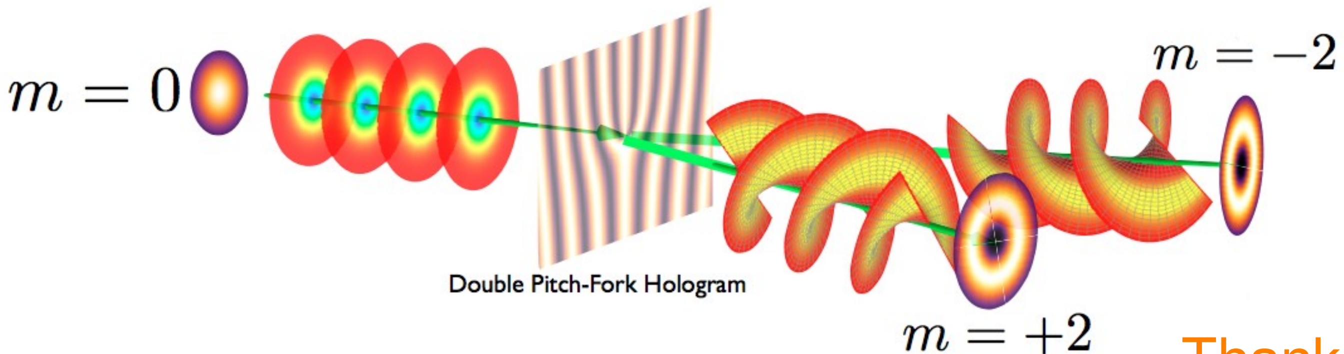
Example: Einstein A and B coefficients for the Fe X spectrum;

LevI-LevF	I- J / Parity	-F	Energy (eV)	Multipol	Gauge	Einstein coefficients A (s^{-1})	gB ($m^3 s^{-2} J^{-1}$)	Oscillator strength GF	Decay width (eV)
1 - 2	1/2 +	1/2 -	3.39446D+01	E1	Babushkin	1.35358D+09	7.92148D+18	5.41457D-02	8.90943D-07
1 - 2	1/2 +	1/2 -	3.39446D+01	E1	Coulomb	1.29696D+09	7.59015D+18	5.18810D-02	8.53678D-07
1 - 1	1/2 +	3/2 -	3.58795D+01	E1	Babushkin	2.94707D+09	1.46045D+19	1.05516D-01	1.93980D-06
1 - 1	1/2 +	3/2 -	3.58795D+01	E1	Coulomb	2.65412D+09	1.31527D+19	9.50275D-02	1.74697D-06
2 - 2	1/2 +	1/2 -	4.66937D+01	E1	Babushkin	5.99420D+06	1.34769D+16	1.26717D-04	3.94546D-09
2 - 2	1/2 +	1/2 -	4.66937D+01	E1	Coulomb	7.32071D+06	1.64593D+16	1.54759D-04	4.81858D-09
2 - 1	1/2 +	3/2 -	4.86286D+01	E1	Babushkin	3.51480D+06	6.99614D+15	6.85074D-05	2.31348D-09
2 - 1	1/2 +	3/2 -	4.86286D+01	E1	Coulomb	4.20990D+06	8.37972D+15	8.20557D-05	2.77101D-09
3 - 2	1/2 +	1/2 -	5.03941D+01	E1	Babushkin	1.70893D+08	3.05647D+17	3.10161D-03	1.12484D-07

Atoms in twisted beams

– generation & control of twisted photons

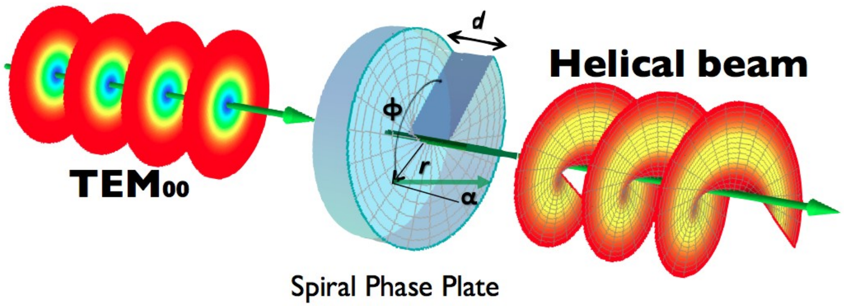


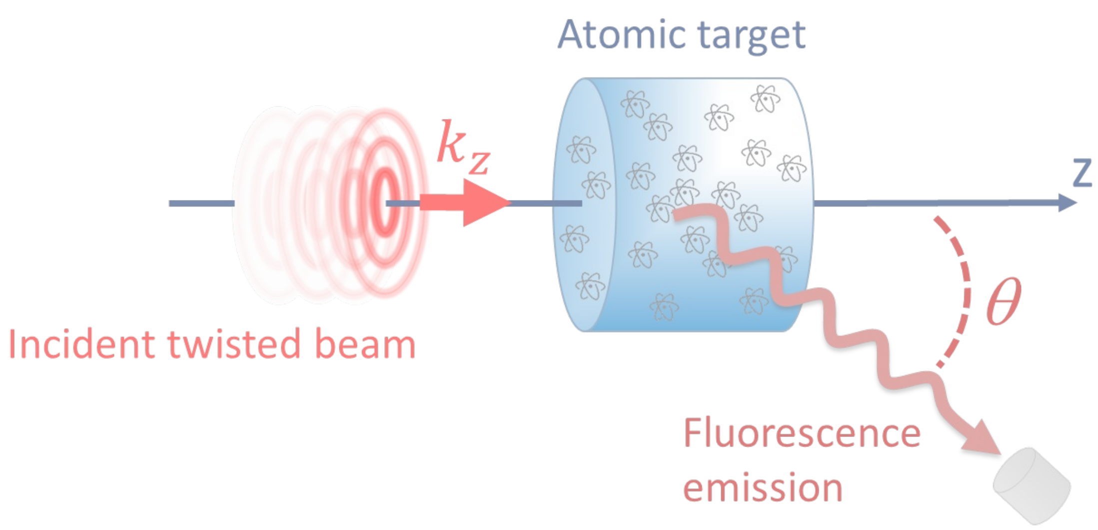
pitch-fork hologram

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

Thanks to recent experimental developments and successes !

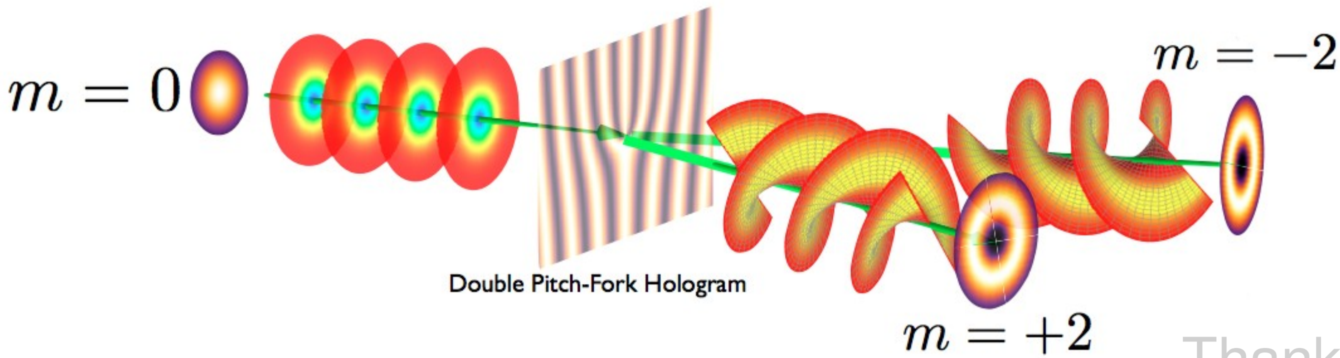
spiral phase plate





Atoms in twisted beams

– excitation and ionization of atomic target (clouds)



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

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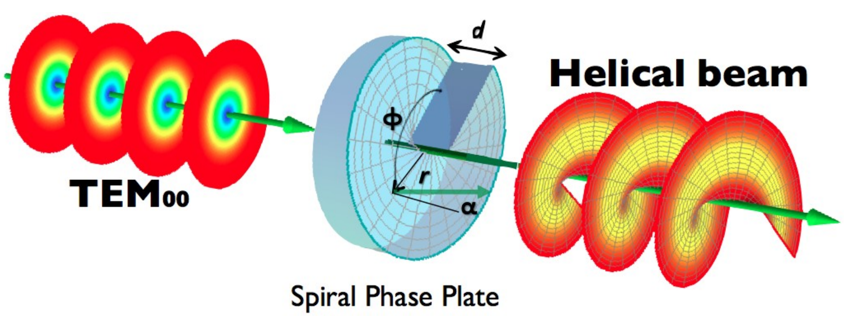
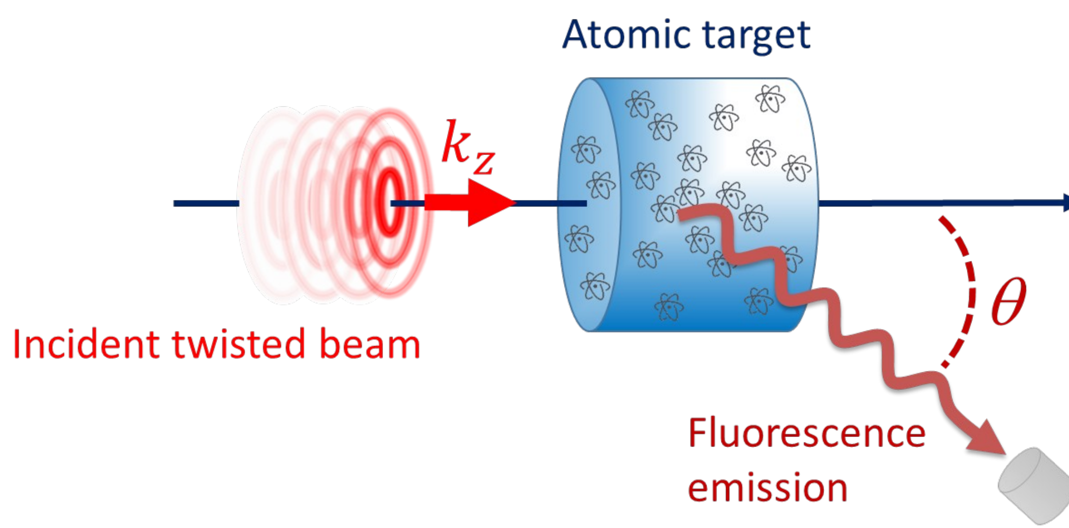
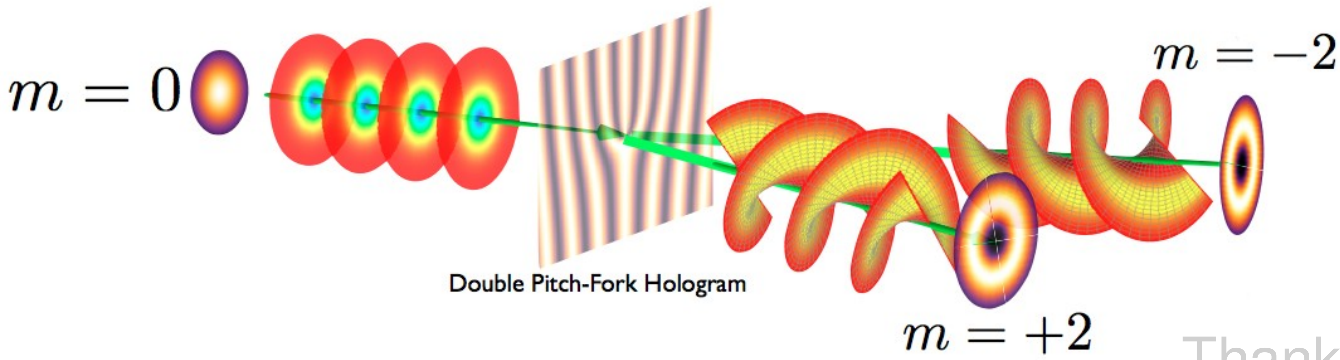




How do these twisted-light beams interact with atomic (and/or molecular) **targets ?**

Atoms in twisted beams

– excitation and ionization of atomic target (clouds)



pitch-fork hologram

Thanks to recent experimental

C. Yao and M. Padgett

spiral phase

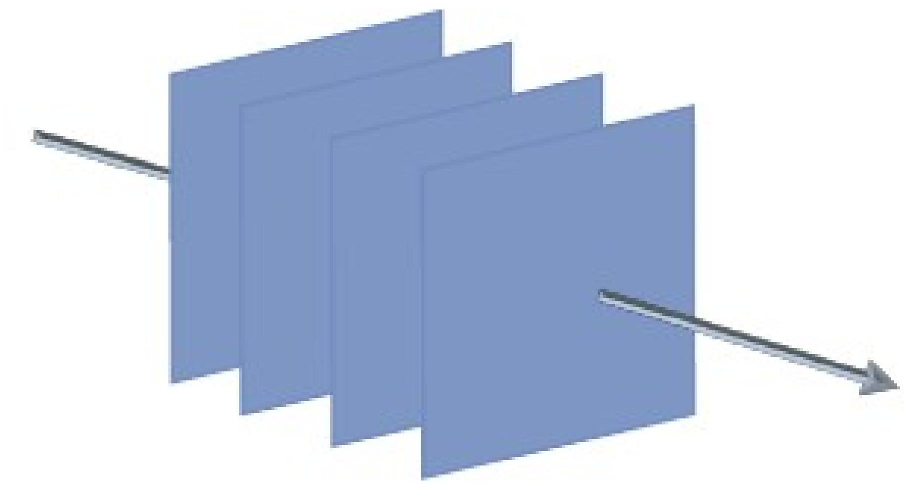
Promising applications of vortex beams:

- Optical tweezer ('single-beam gradient force trap')
- High-bandwidth information encoding in free-space optical communication.
- Higher-dimensional quantum information encoding.
- High-resolution spectroscopy.
- Sensitive optical detection.
- Realization and study of quantum walks.

How do

Plane waves

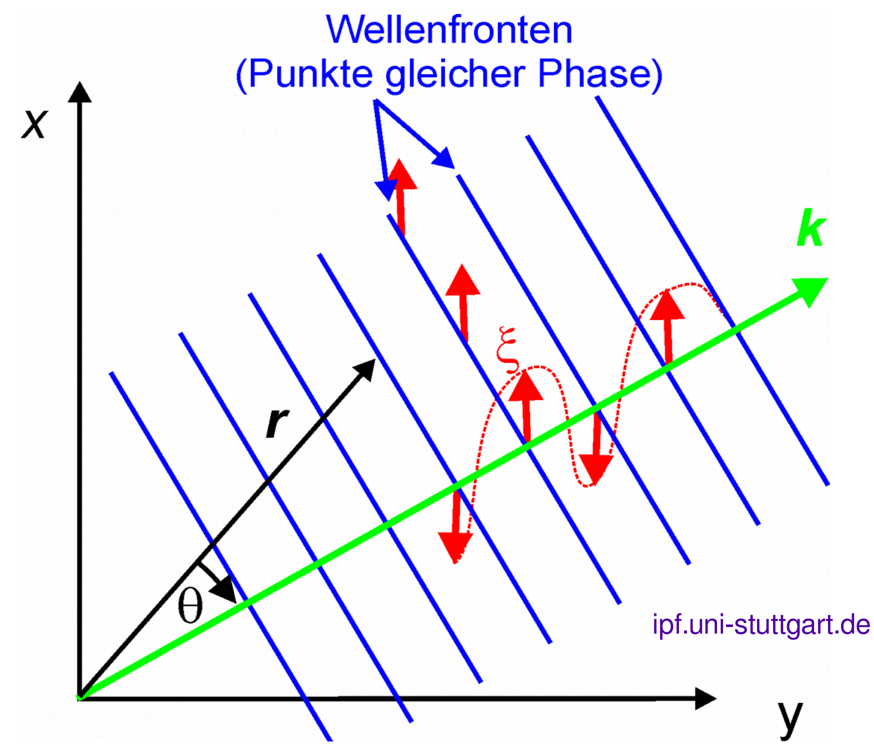
– simple, well-known und frequently applied solutions



- Propagation of electro-magnetic waves;
- in particular, light.
- Free quantum particles
- ...

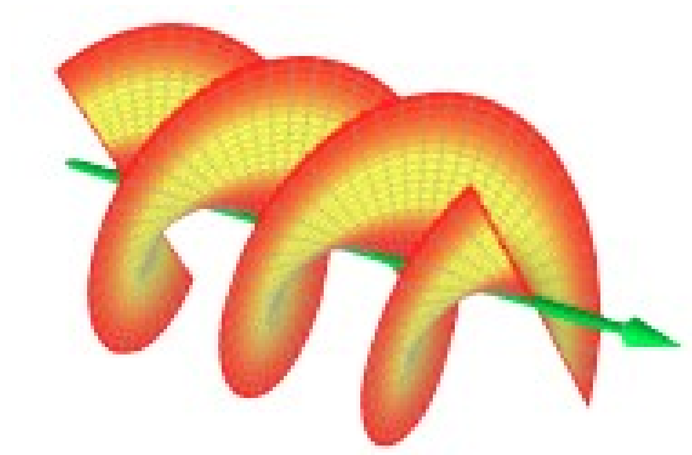
$$\varphi \sim u_\lambda e^{-i\omega t + i k_z z}$$

Quantum numbers: ***k***, λ



Twisted (vortex) beams

– waves with helical wave fronts and orbital angular momentum



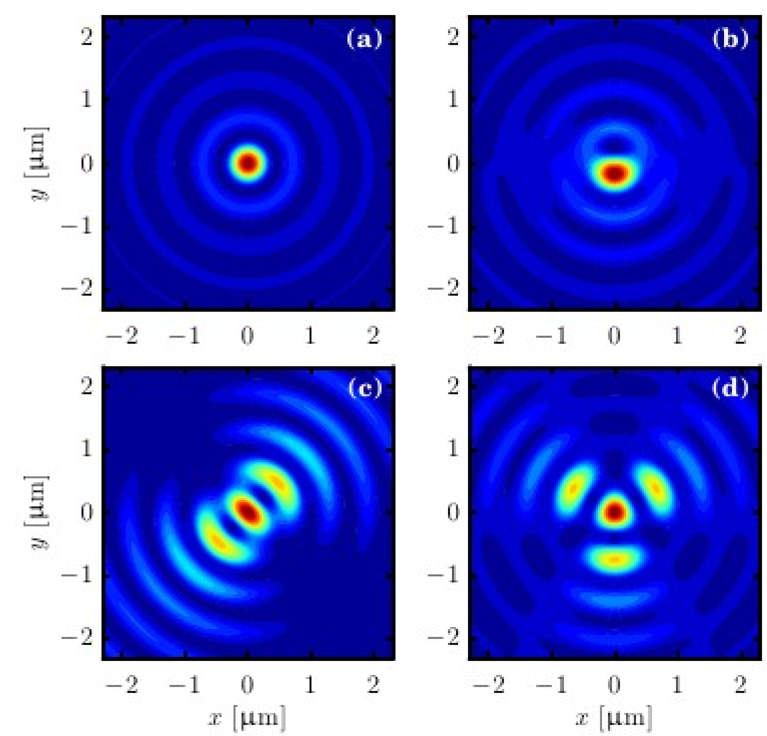
- Laguerre-Gaussian beams
- Bessel beams
- Vector beams

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

Quantum numbers: k_z, k_{perp}, m, λ

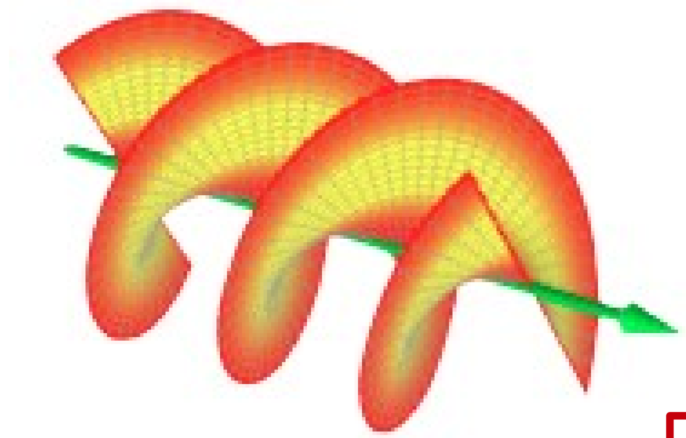
Topological charge, winding number,
projection of OAM, ...

Superposition of Bessel beams



Our 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:

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

Fullfilles Helmholtz's equation.

Probabilities of individual OAM components:

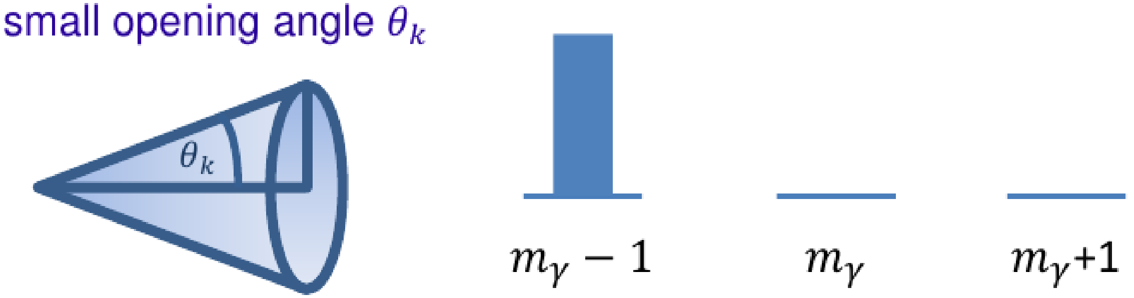
$$P_{m_\gamma - 1} = \frac{1}{4}(\cos\theta_k + \lambda)^2$$

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

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

Bessel beam with small opening angle θ_k

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



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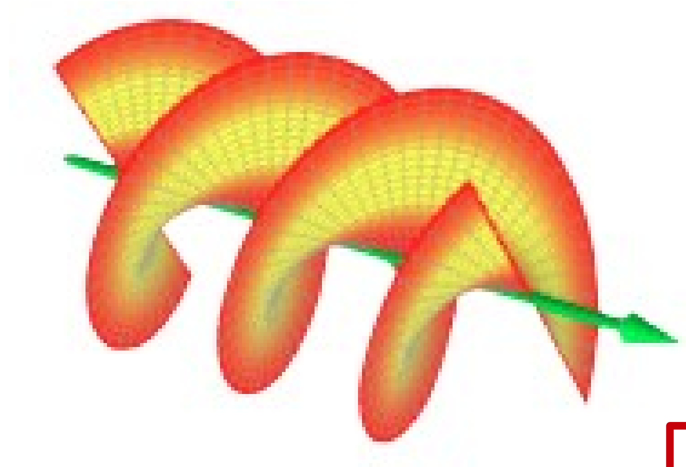
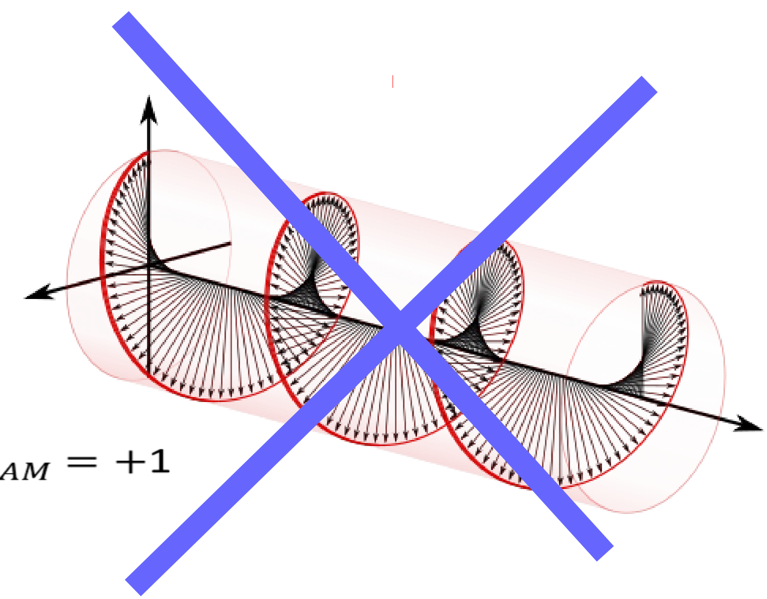


$$\psi \sim e^{-i\omega t + ik_z z}\, e^{im\varphi}\, J_m(k_\perp r)$$

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

Vector potential:

$$\boldsymbol{A}(\boldsymbol{r}) = \int a_{\kappa m}$$



Circularly polarized light

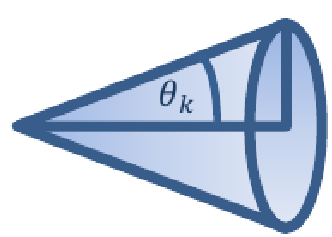
Probabilities of individual OAM compon

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

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

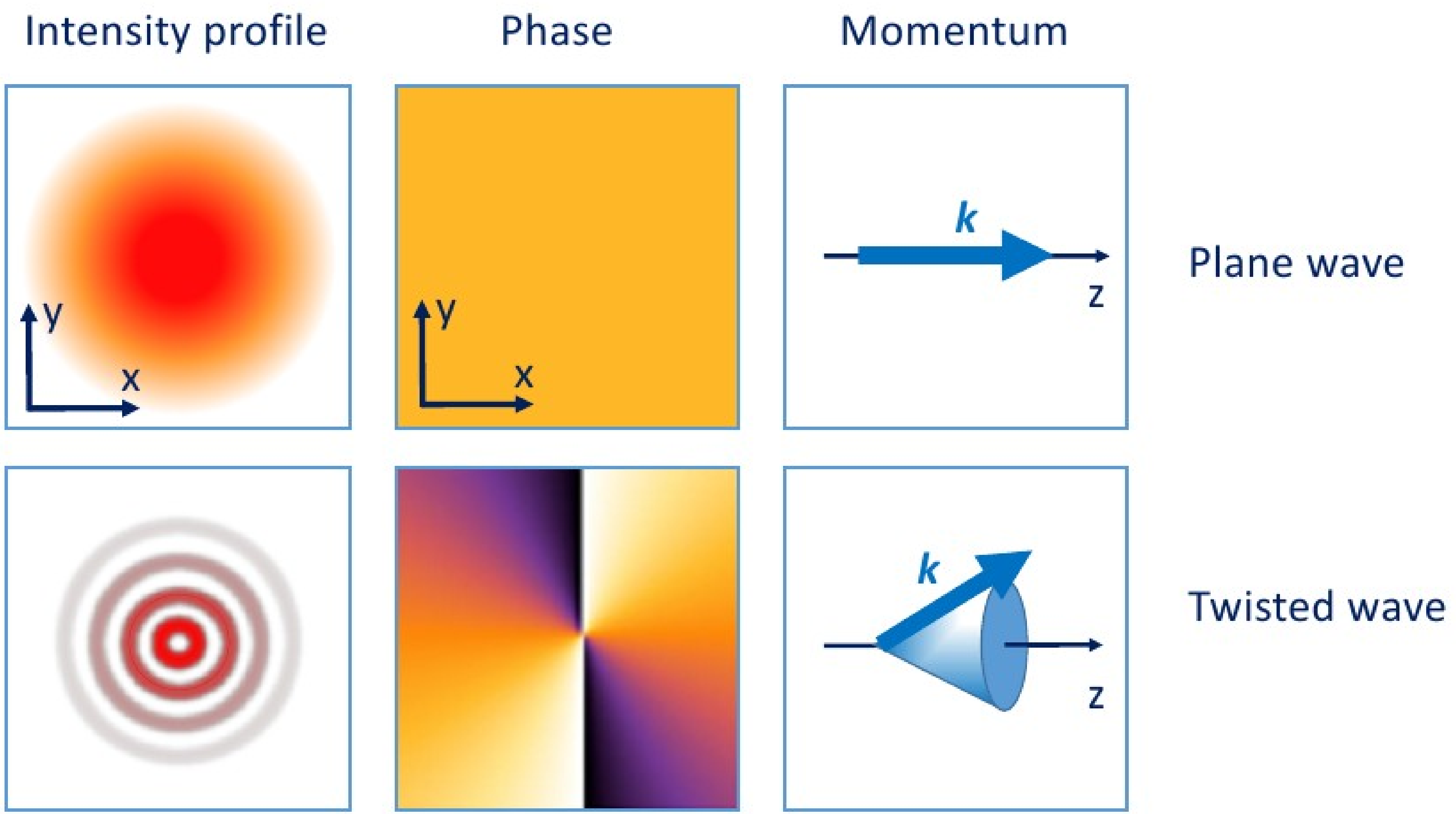
$$P_{m_\gamma + 1} = \frac{1}{4}(\cos\theta_k - \lambda)^2$$

Bessel beam wit
small opening angl



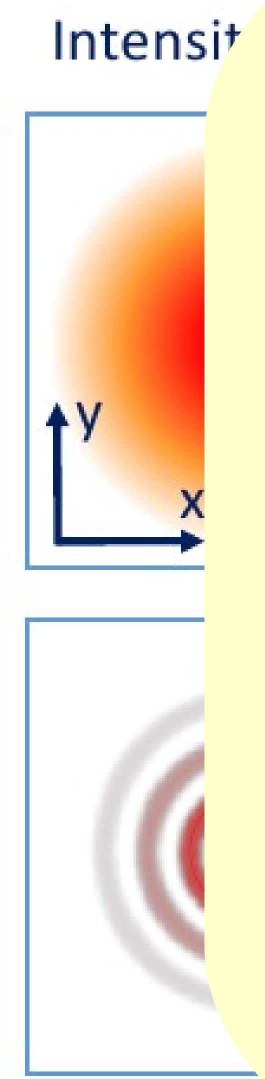
Bessel beams *vs.* plane waves

– representation in position, phase and momentum



Bessel beams *vs.* plane waves

– representation in position, phase and momentum



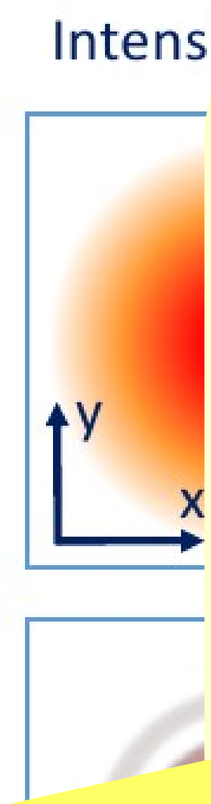
Twisted light

- Twisted photons carry not only spin angular momentum (SAM) but also orbital angular momentum (OAM) along their propagation direction.
- OAM of light implies a spatial distribution of the em field and a phase dependence of the vector potential.
- Wave functions: $e^{im\varphi}$ are eigenfunctions of $L_z = \partial / \partial\varphi$
- SAM and OAM can be separated only in the paraxial approximation.
- Topological charge, m: z-projection of the OAM of the beam.
- A vortex state can propagate freely and does not require any medium nor interaction with other particles and fields to retain its ring-like profile.
- Usable in optical tweezers, high-resolution spectroscopy and high-bandwidth information encoding.

$\rightarrow$ new degree of freedom for light

Bessel beams *vs.* plane waves

– representation in position, phase and momentum



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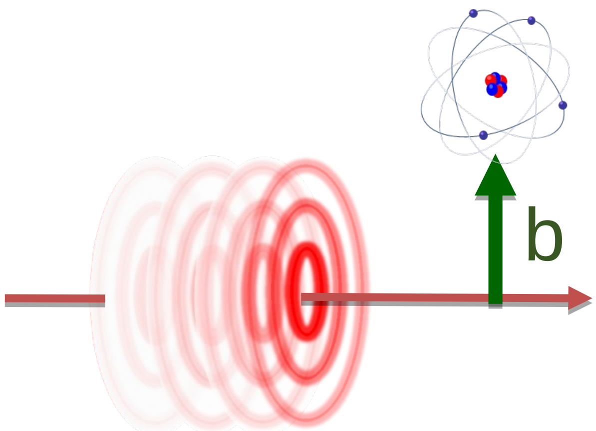
- Topological charg
- A ... nor ... like profile.
- ... ution spectroscopy and high- ... oding.

→ new degree of freedom for light

Until now, (still very) little is known about the interaction of twisted light and beams at the atomic scale.

Photoabsorption of twisted-wave photons

– by atoms with well-defined impact parameter



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

Gives rise to an impact-parameter dependent cross section:

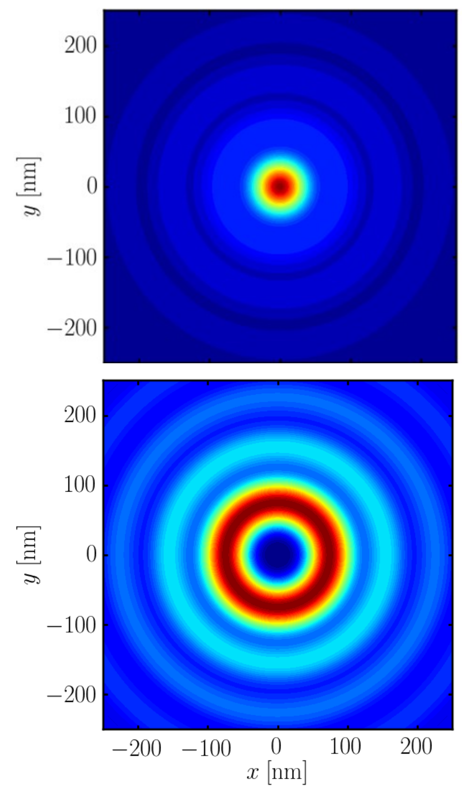
Bessel beam

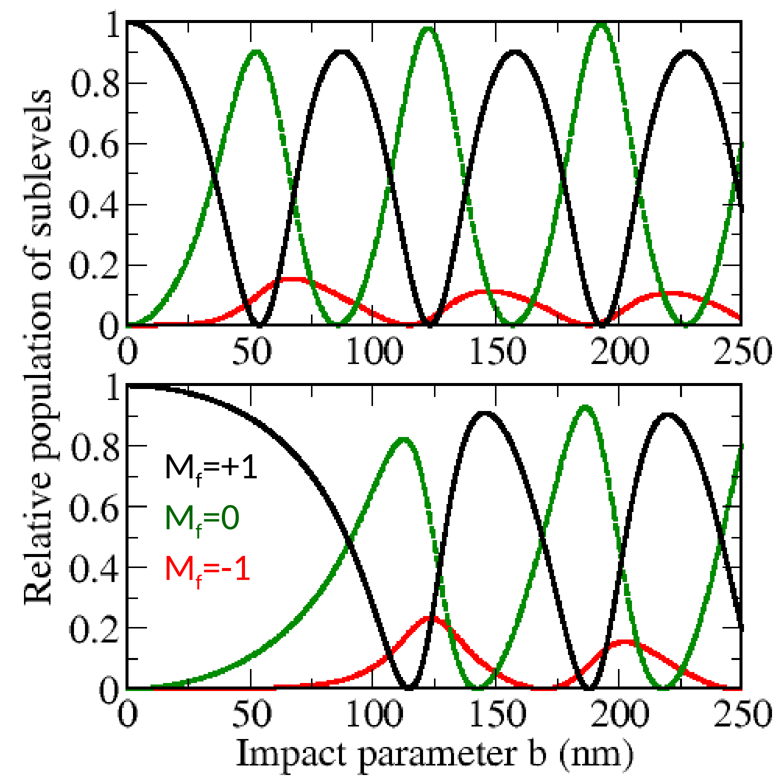
$h\varpi$ = 10 eV, θ = 45°

m_t = 1 and m_t = 3

Characteristic length scale of oscillation

$$b \sim \frac{1}{|k_\perp|}$$





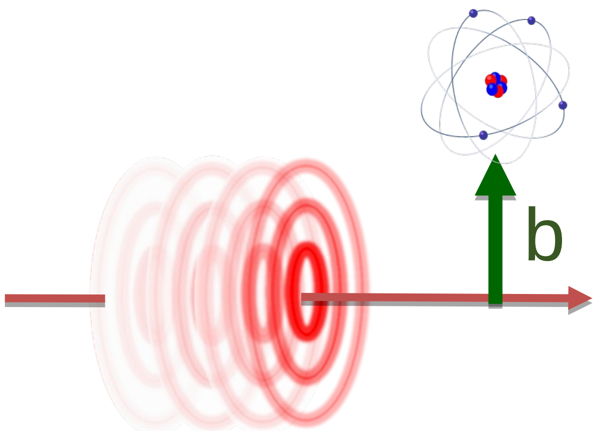
m=1

m=3

A. Surzhykov et al., PRA 90 (2015) 013403.

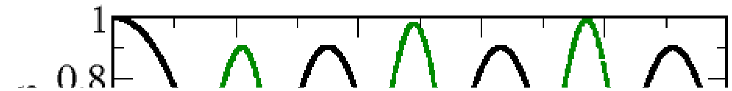
Photoabsorption of **twisted-wave** photons

– by atoms with well-defined impact parameter



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

Gives rise to an impact-parameter dependent cross section:



Bessel beam

$h\varpi$ = 10 eV, θ = 45°

m_t = 1 and m_t = 3

Characteristic length scale of oscillation

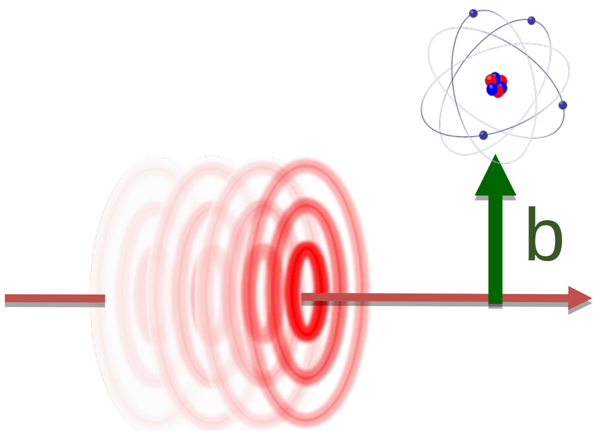
$$b \sim \frac{1}{|k_\perp|}$$

1. Angular distribution of fluorescence
2. Atomic photoionization
3. single vs. mesoscopic vs. macroscopic target
4. radiative recombination (of twisted electrons)
5. inverse Compton effect

...

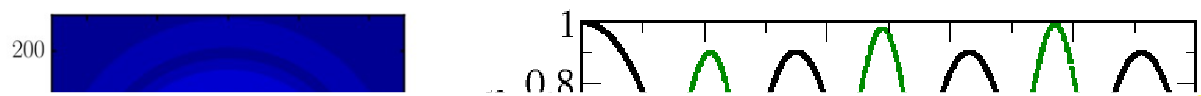
Photoabsorption of **twisted-wave** photons

– by atoms with well-defined impact parameter



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

Gives rise to an impact-parameter dependent cross section:



Bessel beam

$\hbar\varpi$ = 10 eV, θ = 45°

m_t = 1 and m_t = 3

1 Angular distribution of fl...

2 At...

...opic target

...mbination (of twisted electrons)

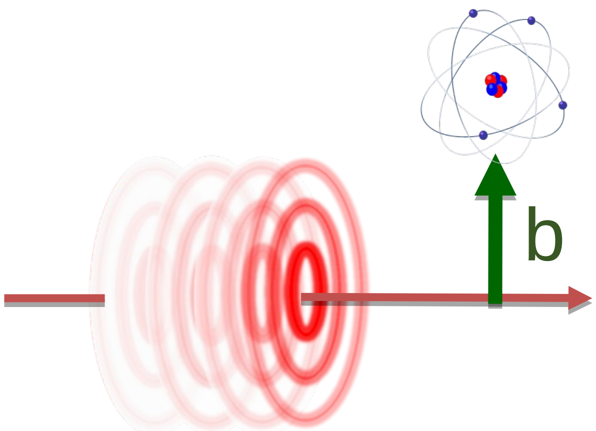
3 inverse Compton effect

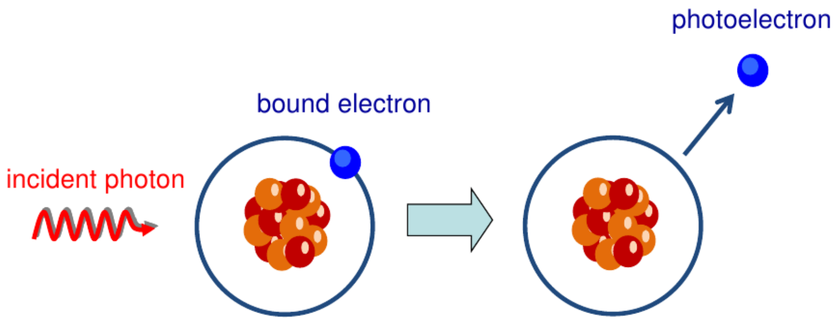
...

Remarkable effects of the „twist" on localized targets;
No dependence on m, if averaged over macroscopic targets.

Photoionization with twisted light

– elementary and very well studied process





Non-relativistic perturbation theory:

$$M^{(\mathrm{pl})}_{m_f m_i}(0,0) = -i\alpha \int \psi^*_{n_f l_f m_f}(\boldsymbol{r})\, e^{ikz} \boldsymbol{\nabla}_\lambda\, \psi_{n_i l_i m_i}(\boldsymbol{r})\, d\boldsymbol{r}$$

Final state in the continuum.

First analysis

- No ϕ_p nor m dependence in the photoelectron spectra if averaged over all impact parameters !
- Only opening angle θ_k matters.
- Reversed viewpoint: Can we make the energy flux in the beams visible ?

Photoionization with twisted light

– How to make the energy flux visible within the beam ?

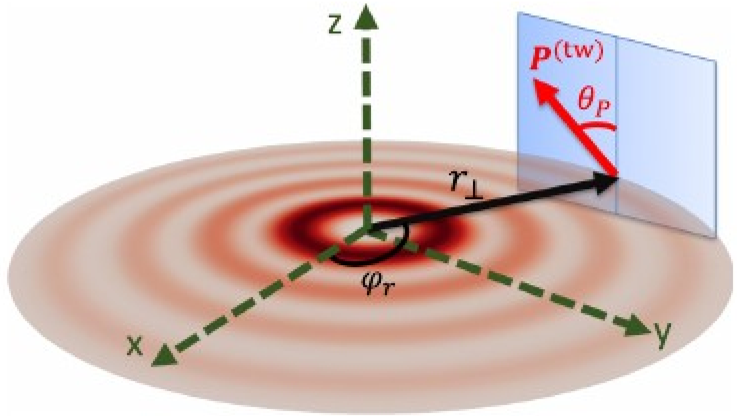
Poynting vector in cylindrical coordinates:

$$\mathbf{P}(\mathbf{r}) = P_{r_\perp}(\mathbf{r})\,\mathbf{e}_{r_\perp} + P_{\varphi_r}(\mathbf{r})\,\mathbf{e}_{\varphi_r} + P_z(\mathbf{r})\,\mathbf{e}_z$$

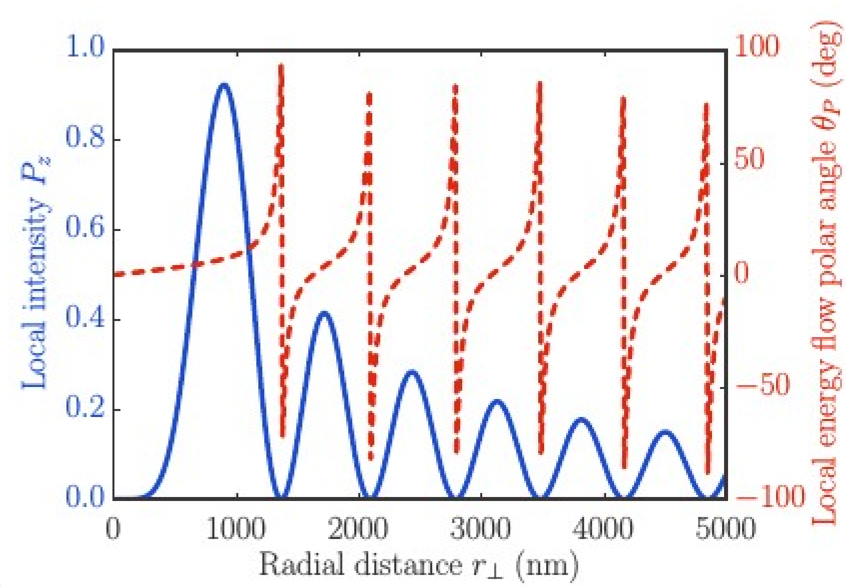
$$P_{r_\perp}(\mathbf{r}) = 0,$$

$$P_{\varphi_r}(\mathbf{r}) = g_P \frac{\varkappa^2}{2\pi} J_{m_\gamma}(\varkappa r_\perp)(c_{+1} J_{m_\gamma - 1}(\varkappa r_\perp) + c_{-1} J_{m_\gamma + 1}(\varkappa r_\perp))$$

$$P_z(\mathbf{r}) = g_P \Lambda \frac{\varkappa k}{2\pi}\left(c_{+1}^2 J_{m_\gamma - 1}^2(\varkappa r_\perp) - c_{-1}^2 J_{m_\gamma + 1}^2(\varkappa r_\perp)\right)$$



Obviously, the Poynting vector depends on the position within the helical wave-front!

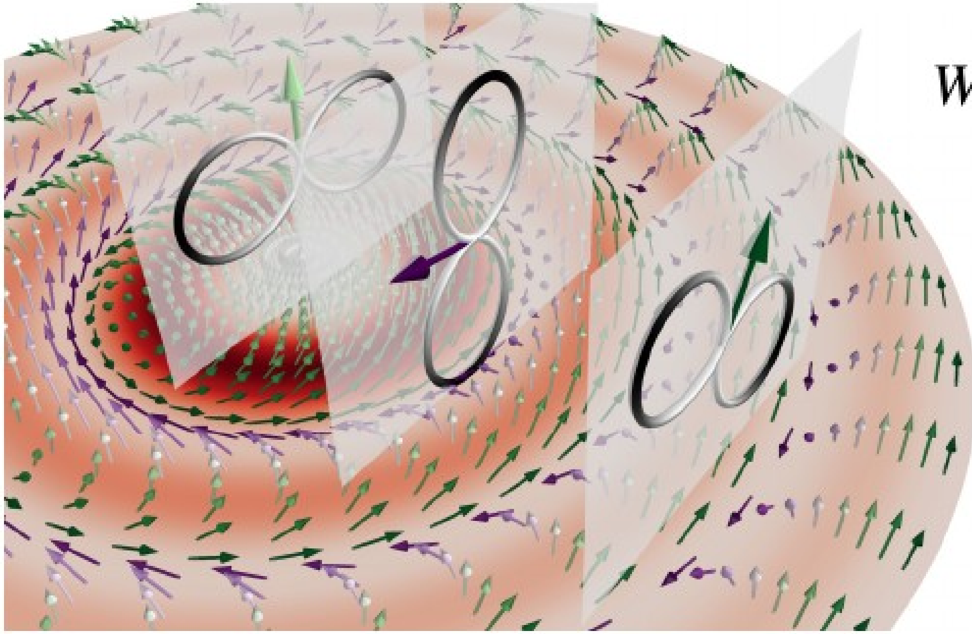


→ How does this dependence of **P** affect the photo-ionization of localized target ?

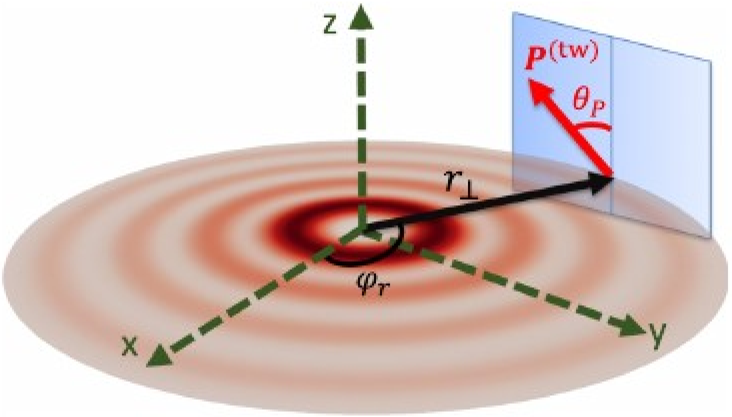
A. Surzhykov et al., PRA 94 (2016) 033420.

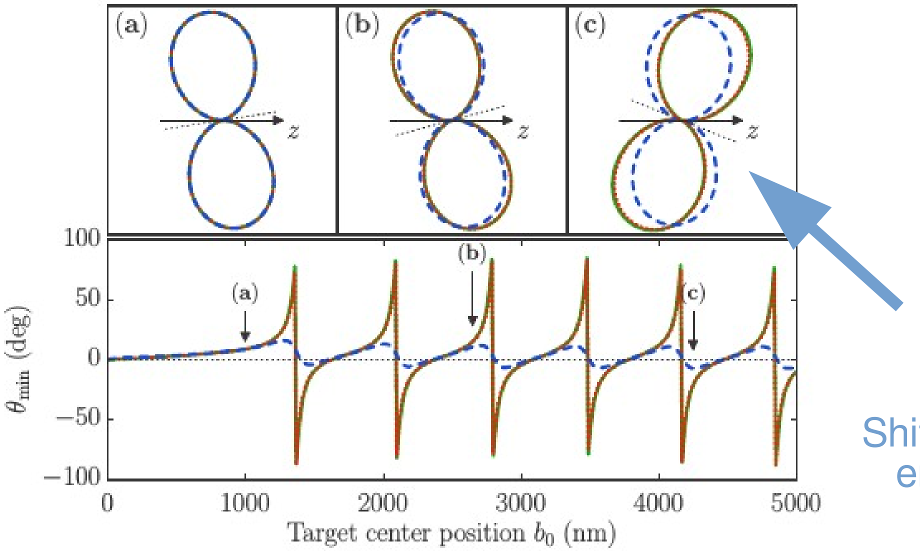
Photoionization with twisted light

– How to make the energy flux visible within the beam ?

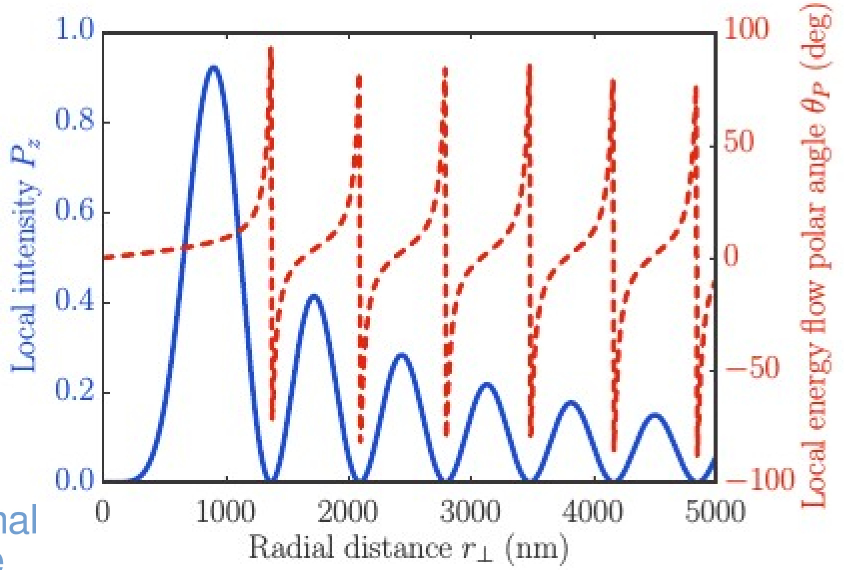


$$W_b^{\rm single}(\theta) = \mathcal{N} \sin^2\left(\theta - \theta_P(b)\right)$$





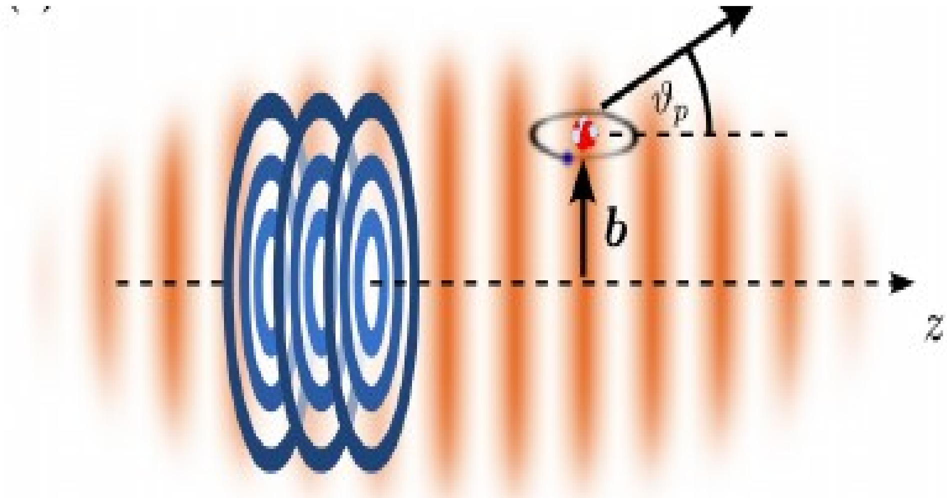
Shift in the minimal emission angle

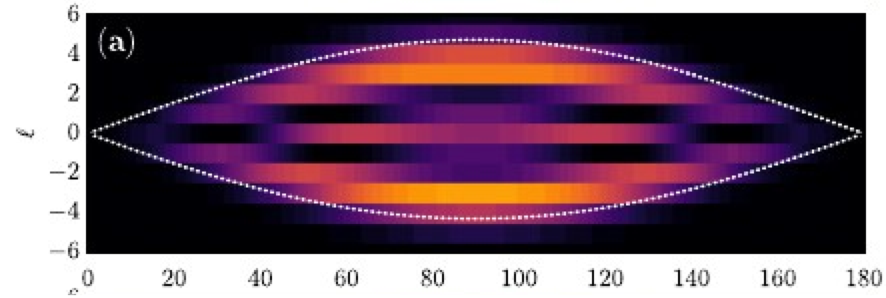


A. Surzhykov et al., PRA 94 (2016) 033420.

Two-color ATI with twisted XUV and intense laser light

– spectral and angular emission of photoelectrons





Amplitude for two-color ATI in strong-field approximation:

$$\mathcal{T}(\boldsymbol{p}) = -i \int_{-\infty}^{\infty} dt \left\langle \Psi^{(V)}_{\boldsymbol{q}(t)} \right| \hat{\boldsymbol{p}} \cdot \boldsymbol{A}_X(\boldsymbol{r}) \left| \phi_0 \right\rangle e^{i\,(E_B - \omega_X)t},$$

Flip of the helicity of laser light !

Ionization probability & circular dichroism:

$$\mathbb{P}^{(\ell)}(\boldsymbol{p}) = |\langle \boldsymbol{p}_\ell | \phi_0 \rangle|^2 \int \frac{d\varphi_k}{2\pi} \, |\mathcal{F}_\ell(\vartheta_k, \varphi_k; \Lambda_X, \Lambda_L)|^2 \quad \longrightarrow \quad \mathrm{CD} = \frac{\mathbb{P}(\boldsymbol{p}; \Lambda_X, \Lambda_L) - \mathbb{P}(\boldsymbol{p}; \Lambda_X, -\Lambda_L)}{\mathbb{P}(\boldsymbol{p}; \Lambda_X, \Lambda_L) + \mathbb{P}(\boldsymbol{p}; \Lambda_X, -\Lambda_L)}$$

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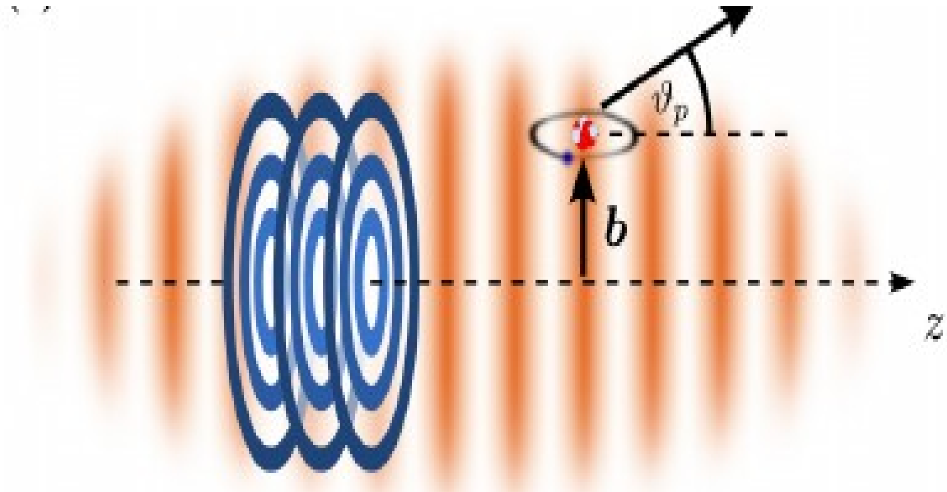

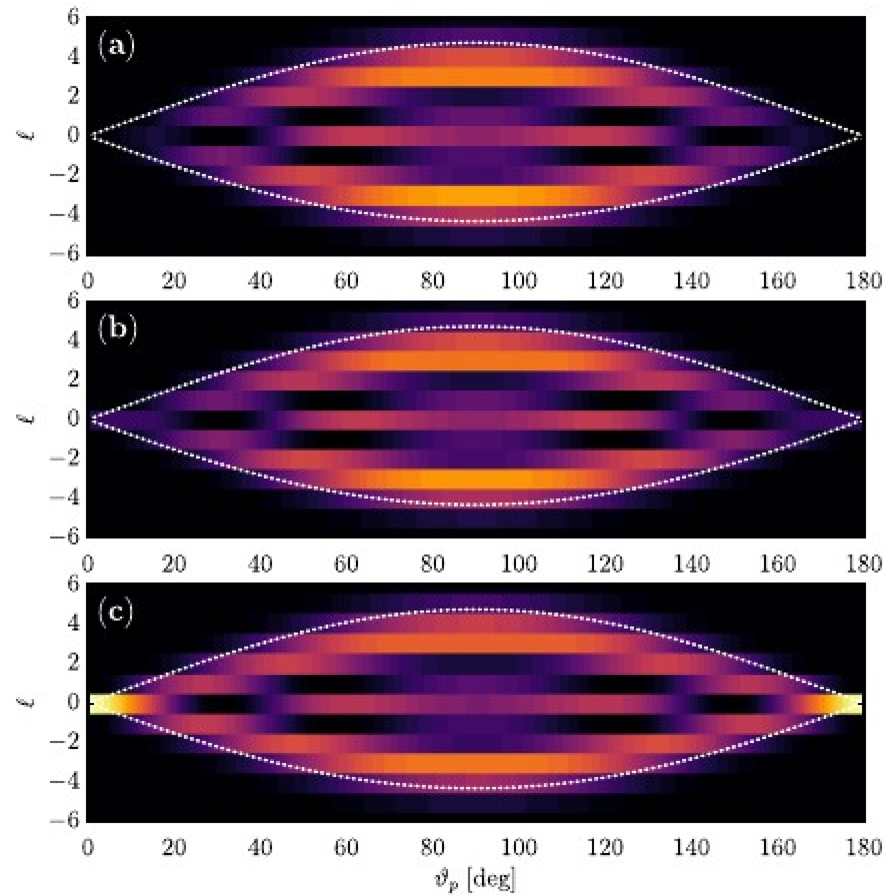

Seven different dichroism signals:

3 magnetic QN → 8 - 1 = 7 ratios

Dichroism due to a flip of ...
the helicity of the assisting NIR laser field.
the helicity of the XUV photons.
the projection of the orbital angular momentum.
the helicity and the orbital angular momentum of the XUV Bessel beam. This is equivalent to just a flip of the projection of the total angular momentum.

D. Seipt et al., PRA 94 (2016) 053420.

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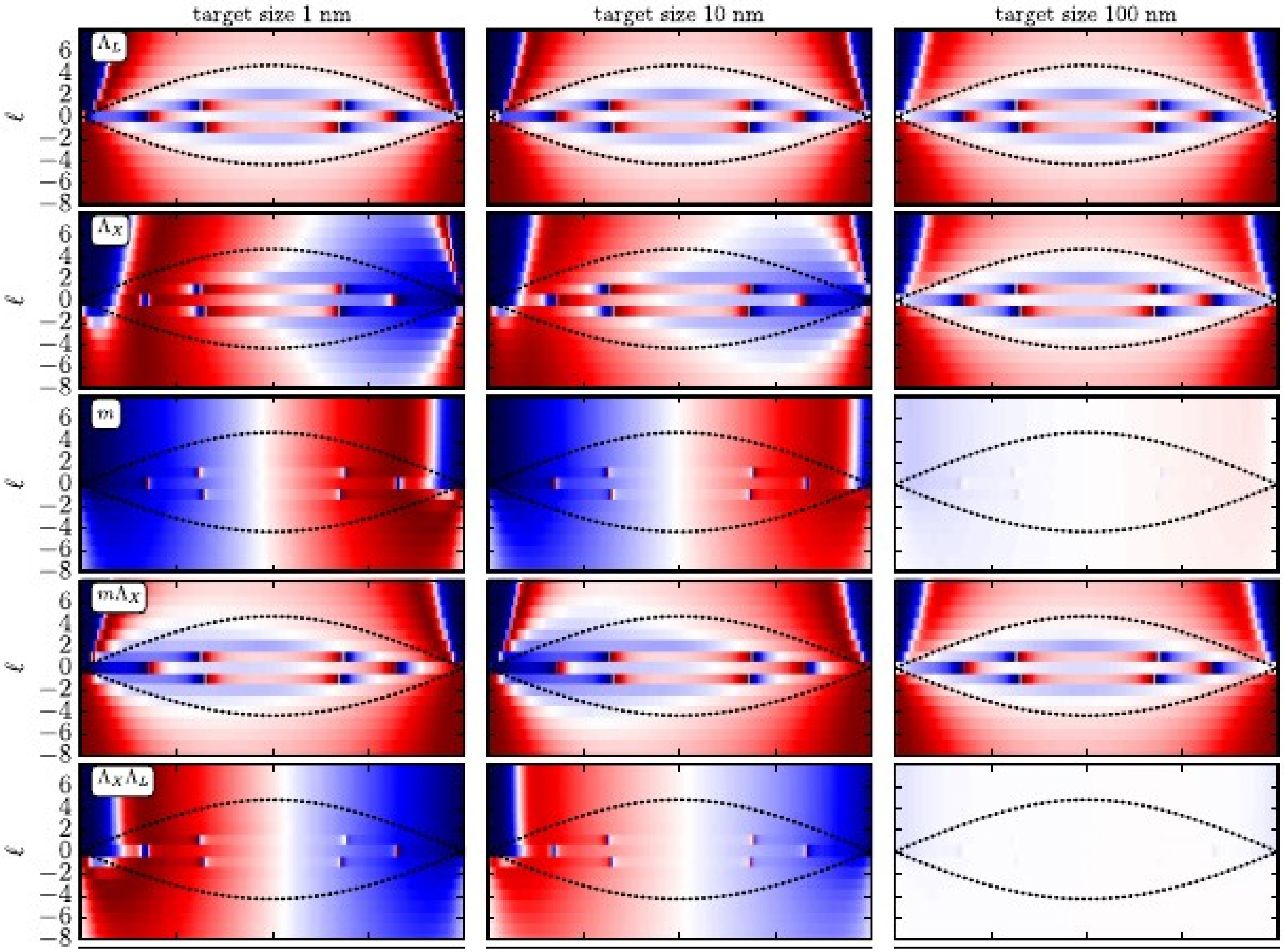


Angular distribution of the sidebands for different target sizes.

Circular dichroism:

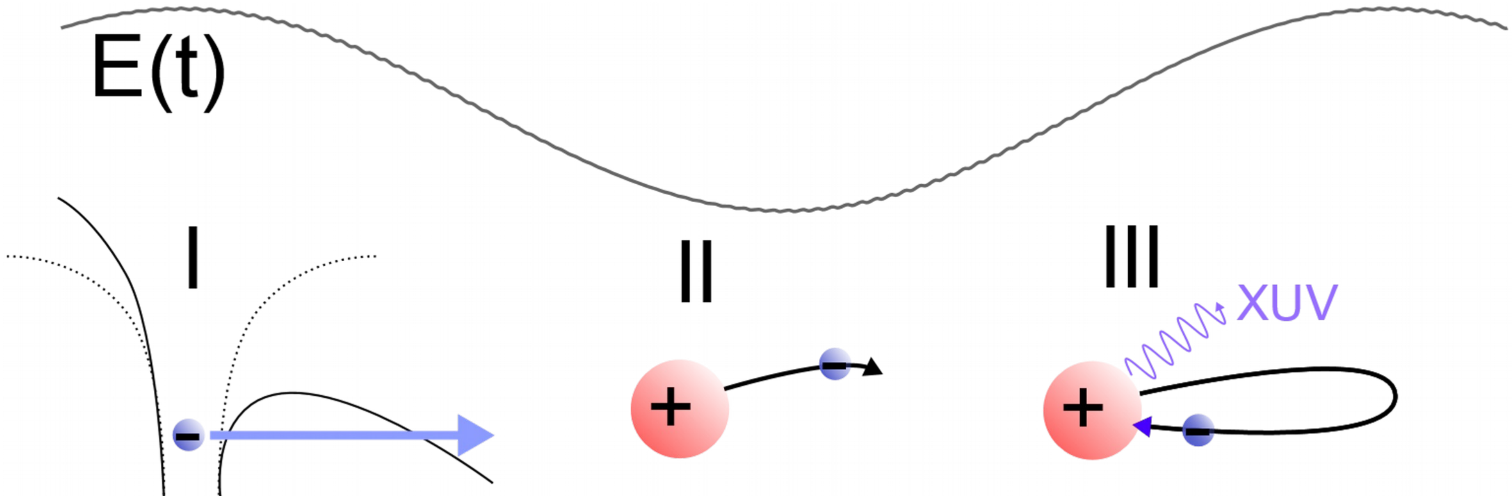
$$\mathrm{CD} = \frac{\mathbb{P}(\boldsymbol{p};\Lambda_X,\Lambda_L) - \mathbb{P}(\boldsymbol{p};\Lambda_X,-\Lambda_L)}{\mathbb{P}(\boldsymbol{p};\Lambda_X,\Lambda_L) + \mathbb{P}(\boldsymbol{p};\Lambda_X,-\Lambda_L)}$$

Again, details of the photon-matter interactions depend on the target size.

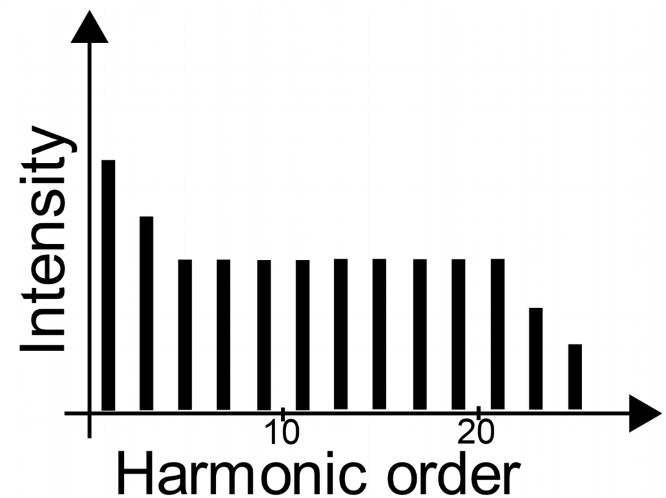
D. Seipt et al., PRA 94 (2016) 053420.

Tailored orbital angular momentum in HHG

– with bi-circular Laguerre-Gaussian beams



I. Ionization
II. Propagation
III. Recombination



P. B. Corkum, PRL **71** (1993) 1994.

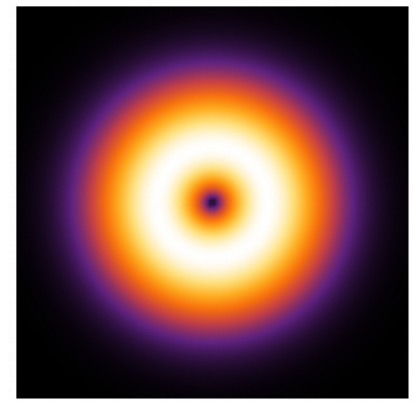
Gaussian *vs.* Laguerre-Gaussian beams

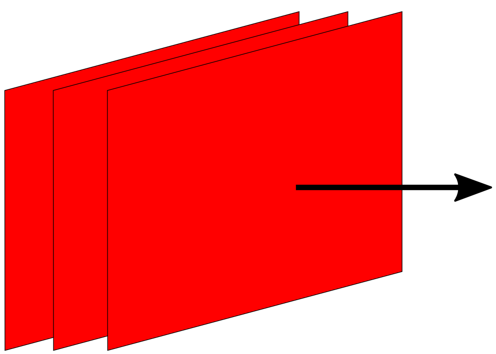
– "twisted" beams with a spatial phase dependence

$$E(r, z) = E_0(r, z)e^{ikz + i\Phi(r,z)}$$

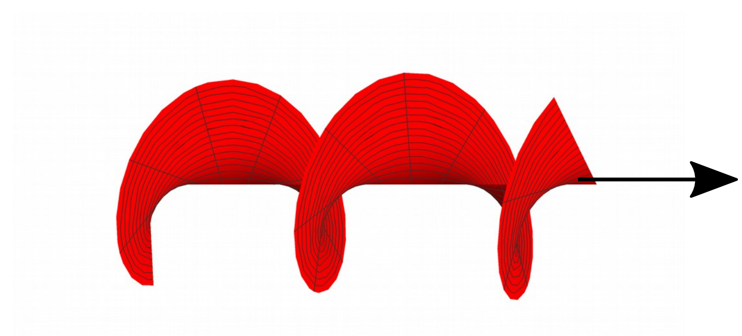
$$E(r, z, \phi) = E_0(r, z)e^{ikz + i\Phi(r,z) + il\phi}$$

Intensity profile



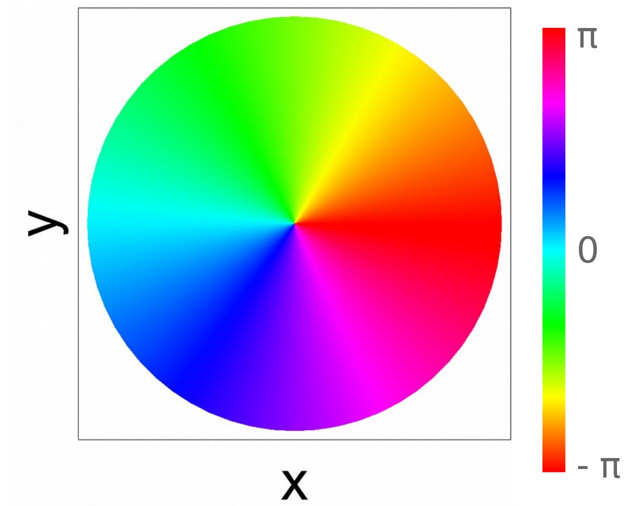


Phase front



HHG with linearly-polarized twisted light

- Only odd H; even harmonics are suppressed.
- Linear scaling of OAM with harmonic order.
- SAM limited by |s| = 1.

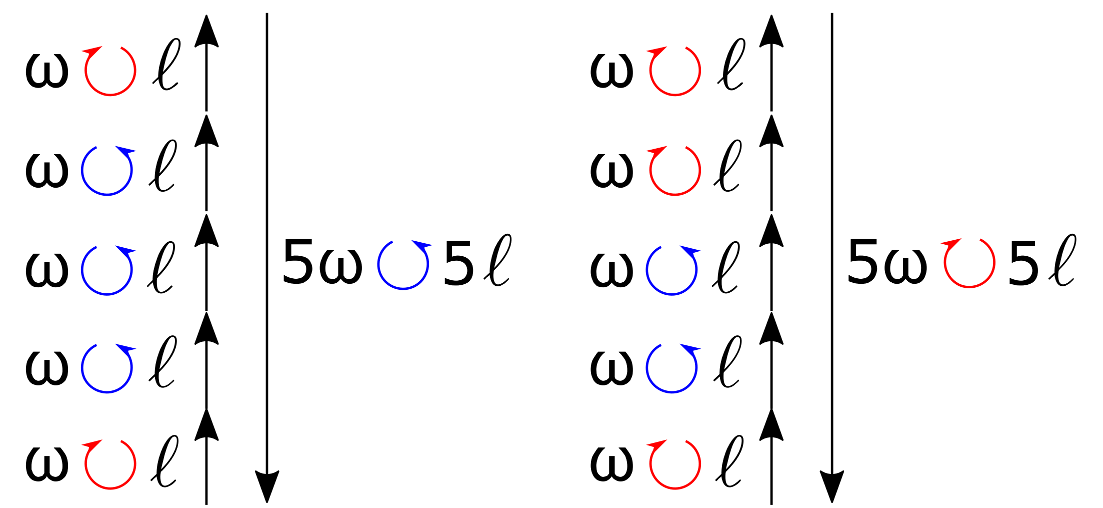


spatial phase dependence

$$LG_{\ell,0}^{\omega} \leftrightarrow \xrightarrow{\text{HHG}} \begin{array}{rcl} \omega_{H_q} & = & q\,\omega \\ \ell_{H_q} & = & q\,\ell \end{array}$$

Linear scaling of OAM

SAM: ±1

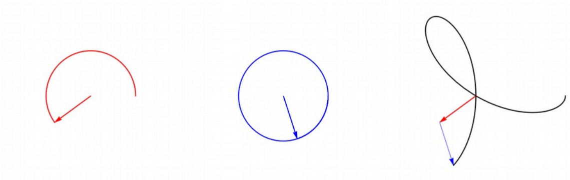


C. Hernandez-Garcia, et al., Phys. Rev. Lett. **111 (2013)** 083602.
G. Gariepy, et al., Phys. Rev. Lett. **113 (2014)**153901.
R. Geneaux, et al., Nature Communications **7** (2016) 12583.

HH are also linearly-polarized ...
as suggested by equal probabilities.

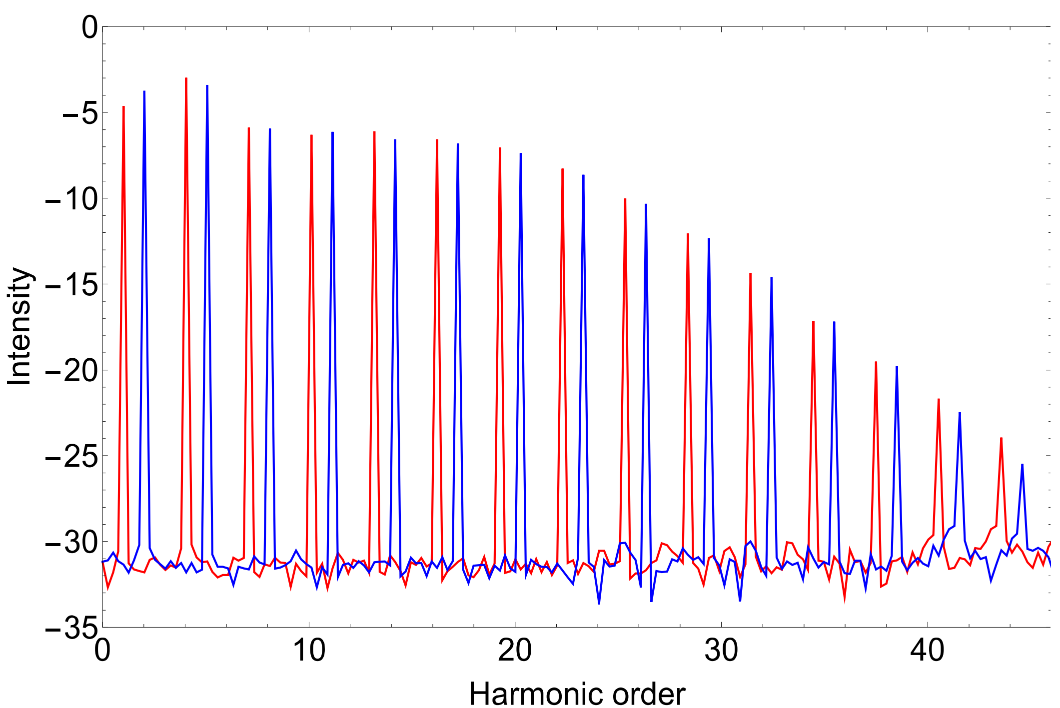
HHG with bi-circular (plane-wave) LG beams

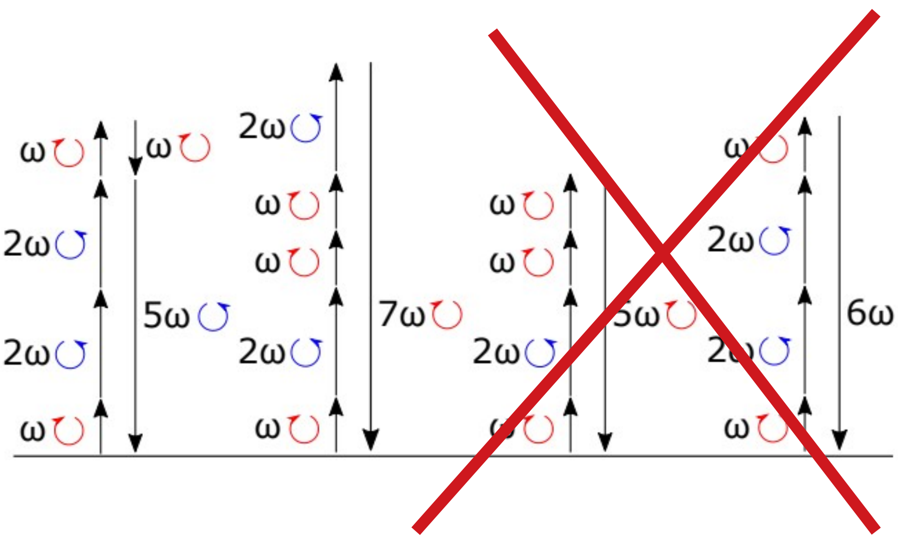
- Superposition of two circularly-polarized fields: $\omega \circlearrowright + 2\omega \circlearrowleft$
- Now, every third harmonic is supressed.
- For each harmonic just one m + n, since SAM = ±1!
- LG with l = 0 refers to a Gaussian beam.



SAM: ±1

$$LG_{0,0}^{\omega \circlearrowright} \oplus LG_{0,0}^{2\omega \circlearrowleft} \xrightarrow{\text{HHG}} \begin{aligned} \omega_{H_q} = q\,\omega &= m\,\omega + n\,2\omega \\ m - n &= \pm 1 \\ \ell_{H_q} &= 0 \end{aligned}$$



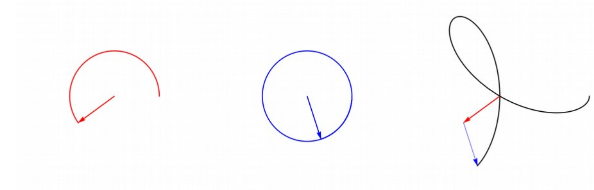


D. B. Milosevic, et al., PRA **61** (2000) 063403 (2000).
K. M. Dorney, et al., PRL **119** (2017) 063201.

HHG with bi-circular (twisted) LG beams

– spatial phase distribution for LG beams with l ≠ 0

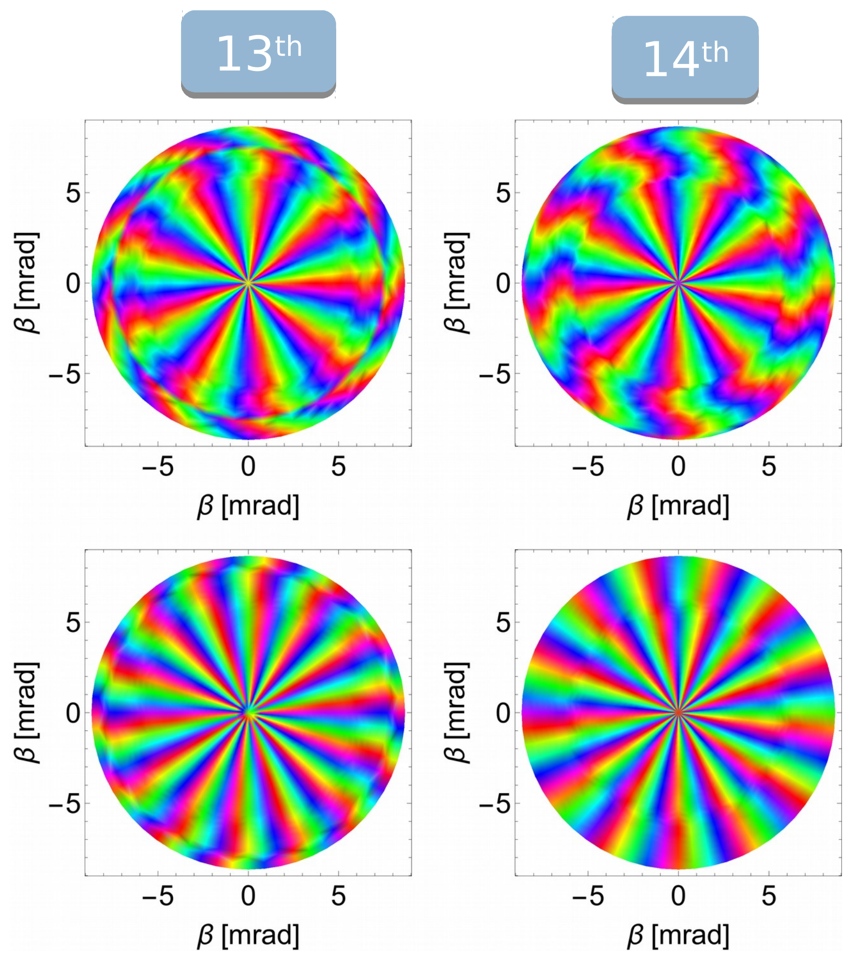
- Superposition of two circular fields: $\omega \circlearrowright + 2\omega \circlearrowleft$
- Now, every third harmonic is supressed.
- What about the OAM of the higher harmonics ?
- No simple scaling of OAM with order of the HHG.



spatial phase distribution in far-field

$$LG_{1,0}^{\omega\,\circlearrowright} \oplus LG_{1,0}^{2\omega\,\circlearrowleft}$$

$$LG_{2,0}^{\omega\,\circlearrowright} \oplus LG_{1,0}^{2\omega\,\circlearrowleft}$$



$$\ell_{H_{13}} = 9 \qquad \ell_{H_{14}} = 9$$

$$\ell_{H_{13}} = 14 \qquad \ell_{H_{14}} = 13$$

W. Paufler et al., PRA **98** (2018) 011401 (R).

OAM of the 13th and 14th harmonic

– simple arithmetics

$$LG_{1,0}^{\omega\circlearrowright} \oplus LG_{1,0}^{2\omega\circlearrowleft} \xrightarrow{\text{HHG}} \begin{aligned} \omega_{H_q} = q\omega &= m\omega + n2\omega \\ m - n &= \pm 1 \\ \ldots ? \end{aligned}$$

SAM: ±1

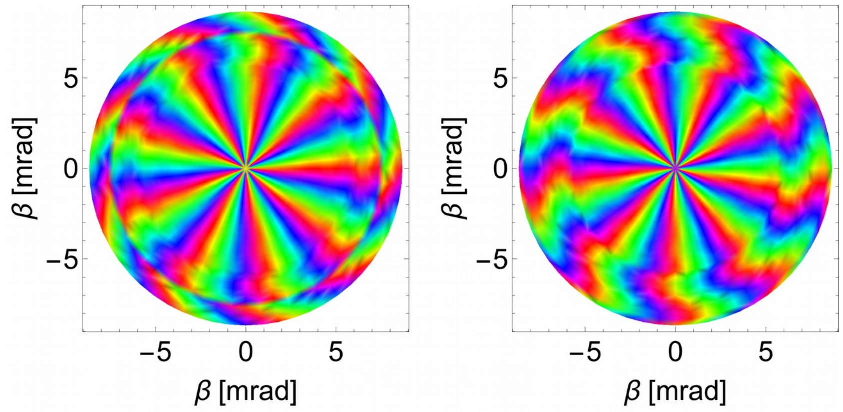
- 13th harmonic $q = 13 \;\Rightarrow\; m = 5 \vee n = 4$

  $\Rightarrow \ell_{H_{13}} = 5 \cdot 1 + 4 \cdot 1 = 9$

- 14th harmonic $q = 14 \;\Rightarrow\; m = 4 \vee n = 5$

  $\Rightarrow \ell_{H_{14}} = 4 \cdot 1 + 5 \cdot 1 = 9$

$$LG_{1,0}^{\omega\circlearrowright} \oplus LG_{1,0}^{2\omega\circlearrowleft}$$



$\ell_{H_{13}} = 9$

$\ell_{H_{14}} = 9$

HHG with bi-circular LG beams: Selection rules

$$r\omega \circlearrowright + s\omega \circlearrowleft$$

$$LG^{\omega \circlearrowright}_{\ell_1,0} \oplus LG^{2\omega \circlearrowleft}_{\ell_2,0} \xrightarrow{\text{HHG}} \begin{array}{rcl} \omega_{H_q} = q\omega & = & m\omega + n2\omega \\ m - n & = & \pm 1 \\ \ell_{H_q} & = & m\,\ell_1 + n\,\ell_2 \end{array}$$

- OAM of each harmonic therefore depends on the OAM of the incident pulses.
- Can we select the OAM of the q^{th} harmonic ?

... **yes:** by choosing the OAM's of the incident fields.

Frequencies	Harmonic order	OAM	m	n	SAM	ℓ_1	ℓ_2
$\omega + 2\omega$	$q = m + 2n$	ℓ_{H_q}	$\frac{q \pm 2}{3}$	$\frac{q \mp 1}{3}$	$m - n = 1$	$\ell_{H_q} + a\,n$	$-\ell_{H_q} - a\,m$
	$q = 1,2,4,5,\ldots$				$m - n = -1$	$-\ell_{H_q} + a\,n$	$\ell_{H_q} - a\,m$
$\omega + 3\omega$	$q = m + 3n$	ℓ_{H_q}	$\frac{q \pm 3}{4}$	$\frac{q \mp 1}{4}$	$m - n = 1$	$\ell_{H_q} + a\,n$	$-\ell_{H_q} - a\,m$
	$q = 1,3,5,7,\ldots$				$m - n = -1$	$-l_{H_q} + a\,n$	$l_{H_q} - a\,m$
$r\omega + s\omega$	$q = rm + sn$	ℓ_{H_q}	$\frac{q \pm s}{r+s}$	$\frac{q \mp r}{r+s}$	$m - n = 1$	$\ell_{H_q} + a\,n$	$-\ell_{H_q} - a\,m$
	$q = r,s,2r+s,2s+r,3r+2s,3s+2r,\ldots$				$m - n = -1$	$-\ell_{H_q} + a\,n$	$\ell_{H_q} - a\,m$

$\rightarrow$ High harmonics with tailored orbital angular momentum.

W. Paufler et al., PRA **98** (2018) 011401 (R).

Summary & Outlook

- Accurate atomic computations are needed for a wide range of applications.
- New experimental facilities require an accurate but still simple handling of (a large number of) levels and amplitudes of different kinds.
- JAC: User-friendly atomic computations of different complexity.
- „Twisted“ photons and beams provide new insights into the elementary light-matter interaction processes.
- This understanding is complementary to the nonlinear and relativistic mechanisms and gives us an alternative route to the control of quantum processes.
- Angular momentum as additional degree of freedom $\rightarrow$ new applications.
- Where shall we go next ??
  - → Rayleigh & Delbrück scattering of twisted light.
  - → Scattering processes at higher intensities.
  - → Selectivity of HHG, phase matching, ...

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