Computation of atomic ionization and recombination properties in synchrotron and FEL radiation

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Universität Oulu und GSI Darmstadt
Moscow, 24th Mai 2012

Thanks to
Dynasty Foundation

Thanks to:

A.N. Grum-Grzhimailo, E. Gryzlova, N.M. Kabachnik, A. Surzhykov (theory)

R. Feifel, M. Meyer, T. Stöhlker and AP@GSI and others (experiment)

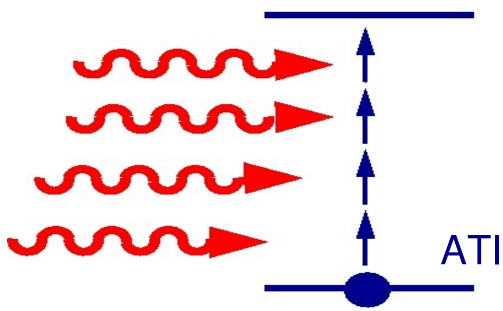
Deutsche Forschungsgemeinschaft DFG

GSI

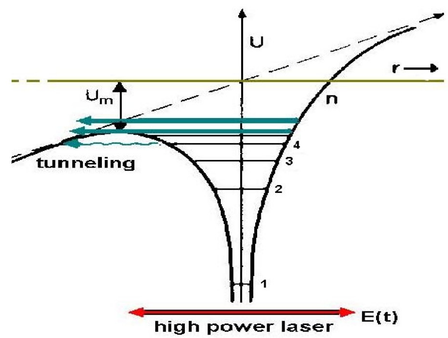
SUOMALAINEN TIEDEAKATEMIA 1908

Computation of atomic ionization and recombination properties in synchrotron and FEL radiation

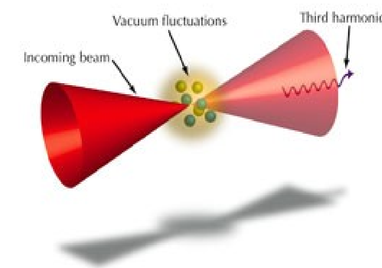
increasing field strength (intensity)



Multiphoton ionization

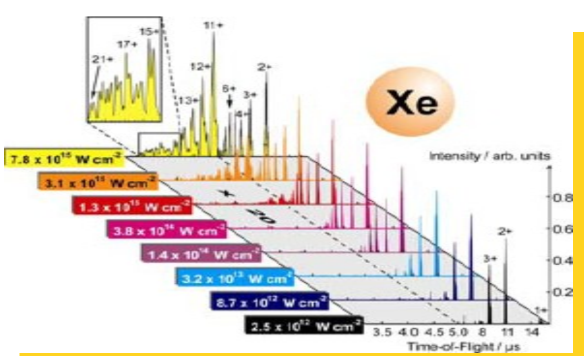


tunnel ionization

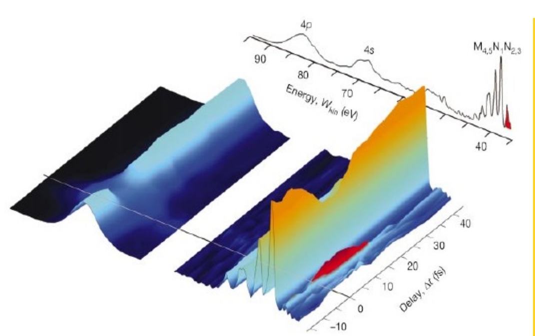


HH and pairs from vacuum

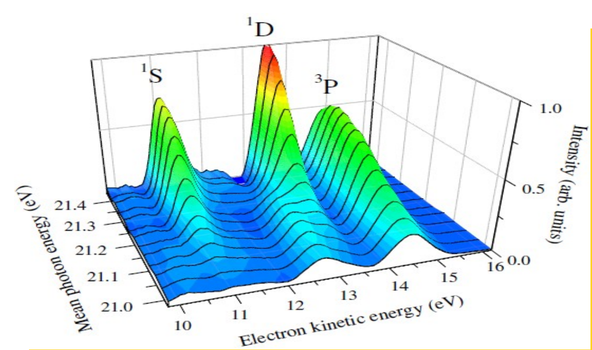
increasing frequency (photon energy)



Multiple ionization



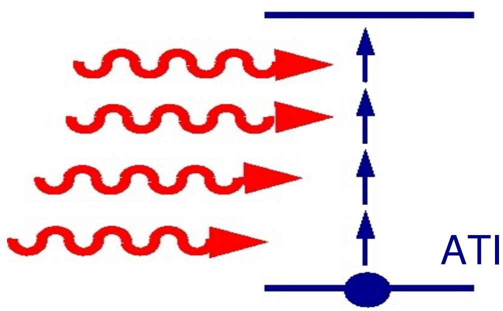
Time-resolved ionization



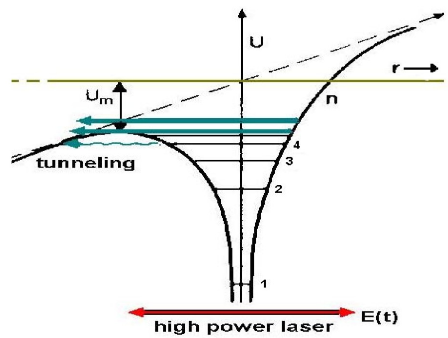
Correlation pattern

Computation of atomic ionization and recombination properties in synchrotron and FEL radiation

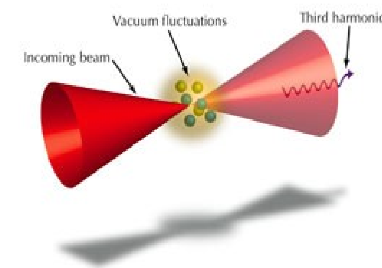
increasing field strength (intensity)



Multiphoton ionization



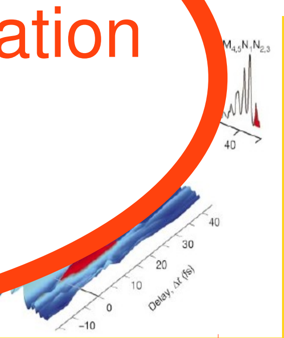
tunnel ionization

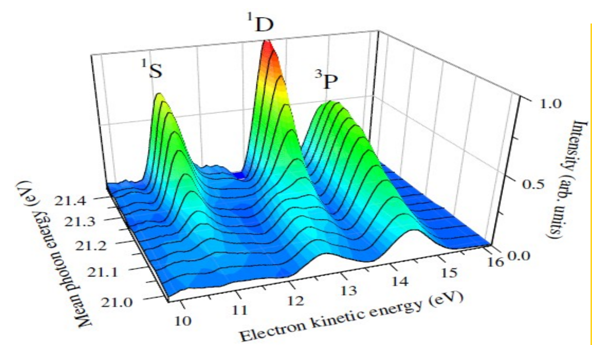


HH and pairs from vacuum

(photon energy)

Atomic double & multiple ionization
@ synchrotrons and FEL





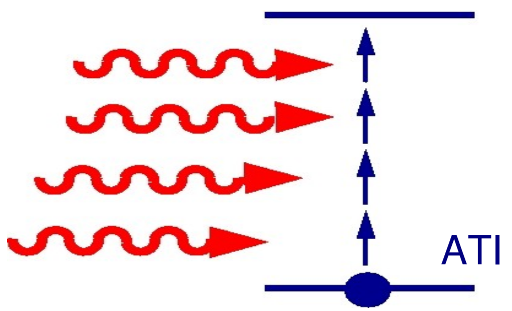
Multiple ionization

Time-resolved ionization

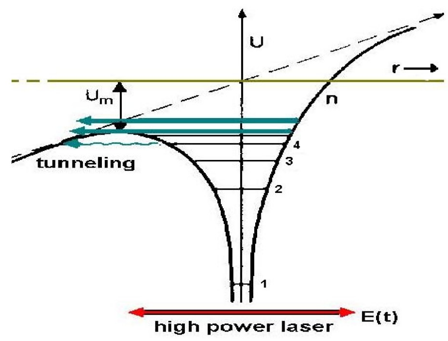
Correlation pattern

Computation of atomic ionization and recombination properties in synchrotron and FEL radiation

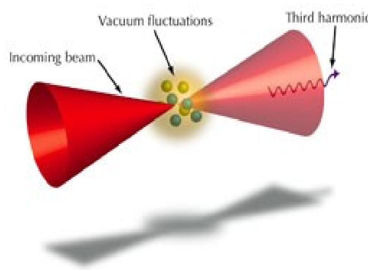
increasing field strength (intensity)



Multiphoton ionization



tunnel ionization



HH and pairs from vacuum

Ato

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Plan of this talk

I. Recombination: Exploring the elementary interactions in strong fields

II. RATIP: Tools for computations

III. Weak-fields: Multiple ionization of noble gases

IV. Ionization dynamics in intense FEL pulses; summary

Exploring the elementary interactions in strong fields

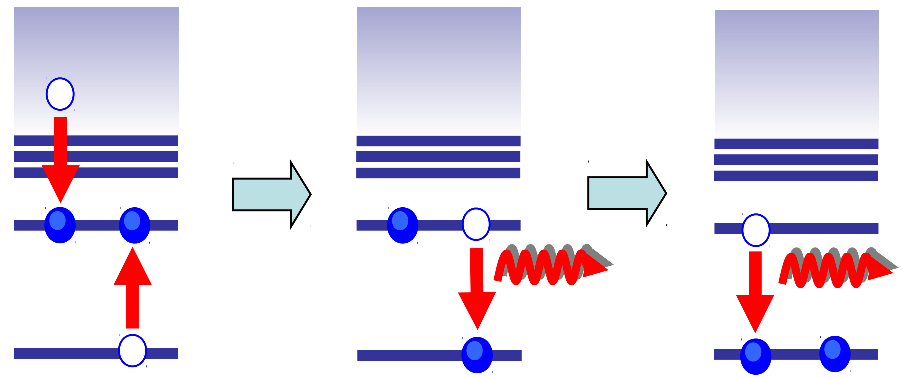
– electron-photon vs. electron-electron interactions

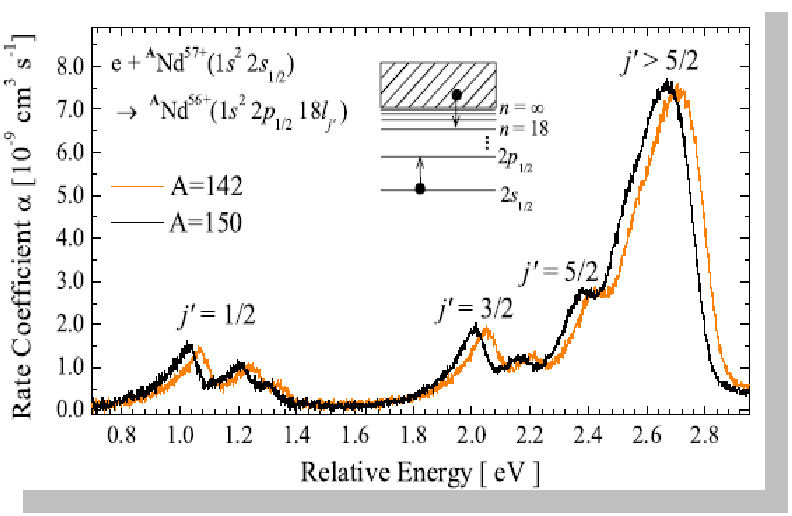
Photoionization

Autoionization

Radiative electron capture (REC)

Dielectronic recombination (DR)





C. Brandau et al., PRL 100 (2008) 073201; PRL 89 (2002) 053201; PRL 91 (2003) 073202

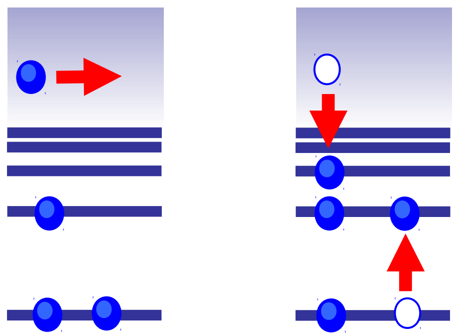
- Dielectronic recombination (DR) process provides a unique tool for precise spectroscopy of HCI and, especially, doubly excited ionic states.
- Sensitive to nuclear, isotope and QED properties

- Interactions in intense (Coulomb) fields
- Great importance for plasma and FEL physics.

Exploring the elementary interactions in strong fields

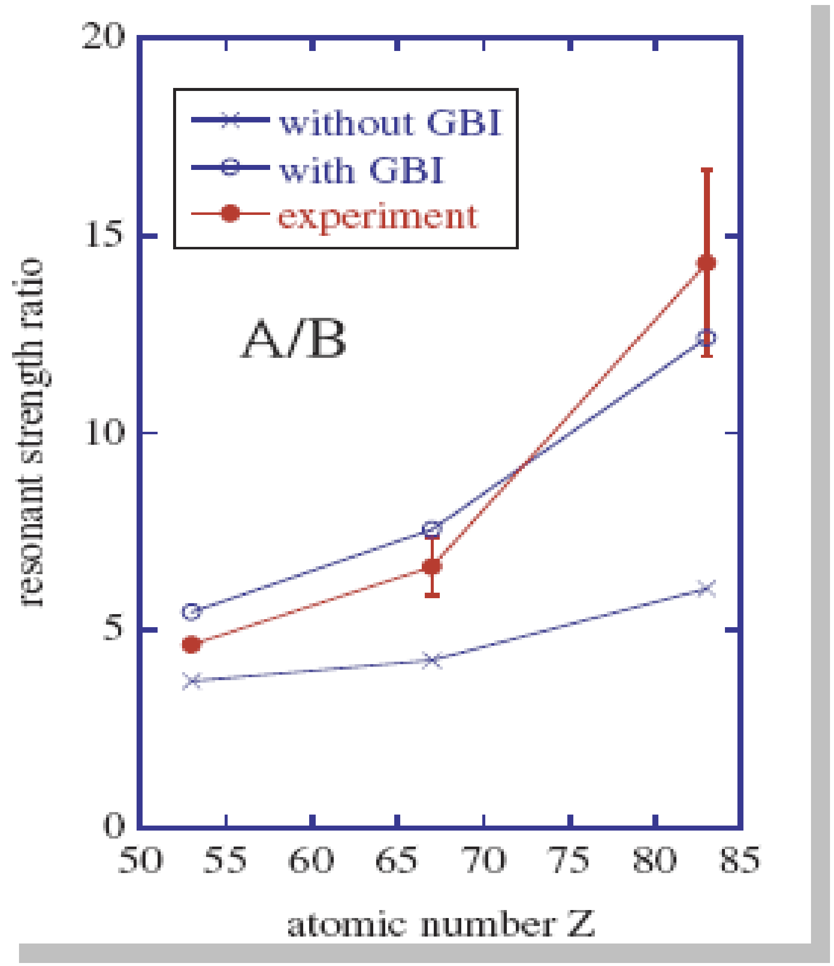
– finger prints upon magnetic and retarded interactions

K-LL DR into initially lithium-like ions:



$$\left[1s^2\ 2s\right]_{1/2} + e^- \rightarrow \left[1s\ 2s^2\ 2p\right]_1$$

relative to the the
1s 2s 2p^2 J=1
resonance



$$V_{ee} = V^C + V^B = \frac{1}{r_{12}}$$

$$+\left(-\alpha_1\alpha_2\frac{\cos\omega r_{12}}{r_{12}} + (\alpha_1\nabla_1)(\alpha_2\nabla_2)\frac{\cos\omega r_{12}}{\omega^2 r_{12}}\right)$$

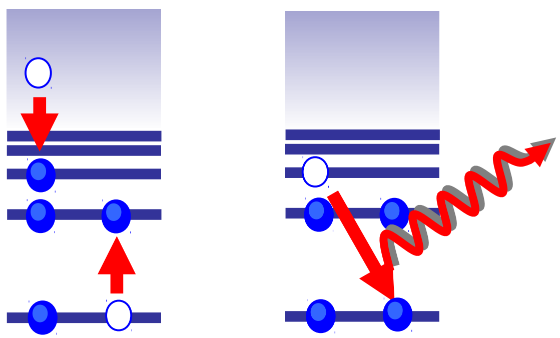
Breit interaction

EBIT measurements:
N. Nakamura et al., PRL 100 (2008) 073203.

Exploring the elementary interactions in strong fields

– finger prints upon magnetic and retarded interactions

K-LL DR into initially lithium-like ions:

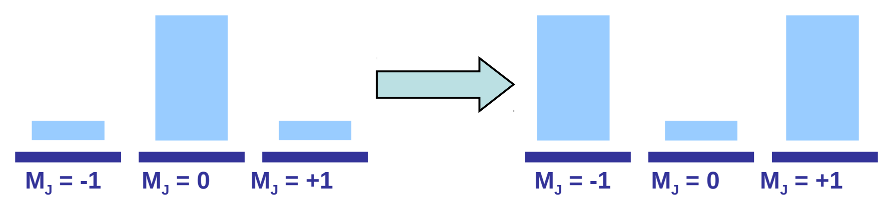


$$\left[1s\ 2s^2\ 2p\right]_1 \Rightarrow \left[1s^2\ 2s^2\right]_0 + \gamma$$

$$V_{ee} = V^C + V^B = \frac{1}{r_{12}}$$

$$+\left(-\alpha_1 \alpha_2 \frac{\cos \omega r_{12}}{r_{12}} + (\alpha_1 \nabla_1)(\alpha_2 \nabla_2) \frac{\cos \omega r_{12}}{\omega^2 r_{12}}\right)$$

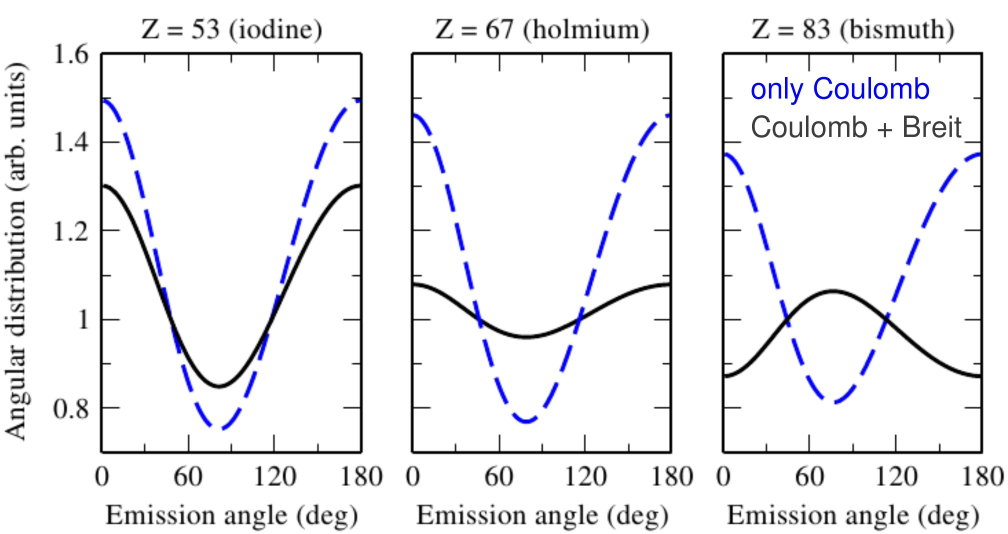
only Coulomb → Coulom + Breit



A_2 = -0.516 A_2 = +0.374

$$W(\theta) \propto 1 + \frac{A_2}{\sqrt{2}} P_2(\cos\theta)$$

Angular distribution of emitted photons

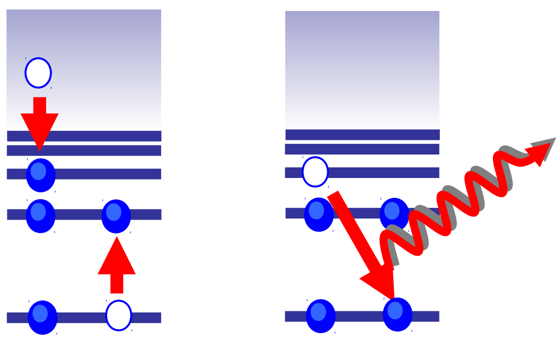


S. Fritzsche et al., PRL 103 (2009) 113001.

Exploring the elementary interactions in strong fields

– finger prints upon magnetic and retarded interactions

K-LL DR into initially lithium-like ions:



$$\left[1s\ 2s^2\ 2p\right]_1 \Rightarrow \left[1s^2\ 2s^2\right]_0 + \gamma$$

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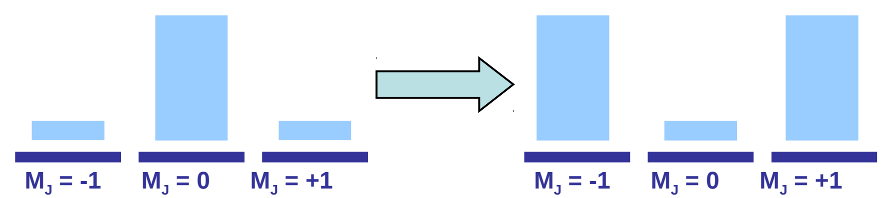
$$+\left(-\alpha_1\alpha_2\frac{\cos\omega r_{12}}{r_{12}} + (\alpha_1\nabla_1)(\alpha_2\nabla_2)\frac{\cos\omega r_{12}}{\omega^2 r_{12}}\right)$$

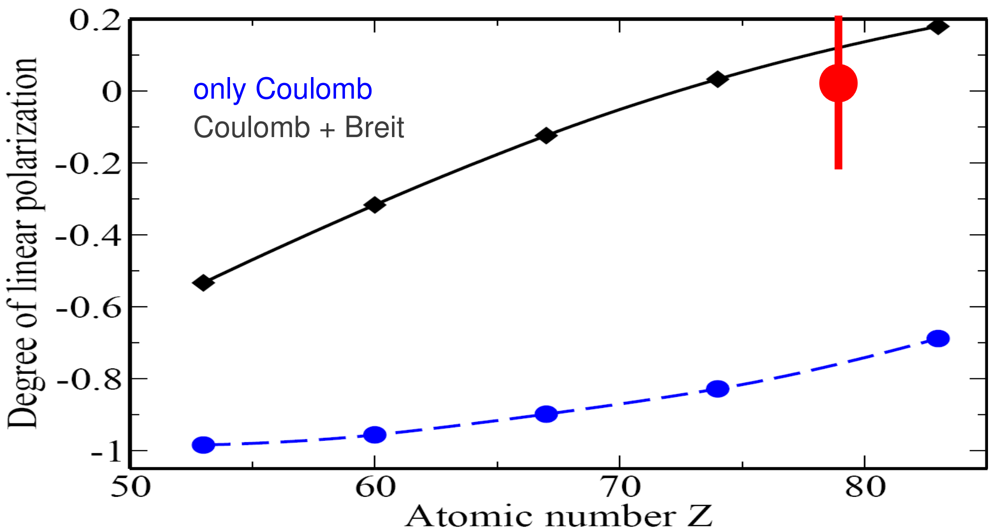
only Coulomb → **Coulom + Breit**



A_2 = -0.516 A_2 = +0.374

$$P(\theta = \pi/2) = \frac{-3\sqrt{2}A_2}{4-\sqrt{2}A_2}$$

Linear polarization of emitted photons

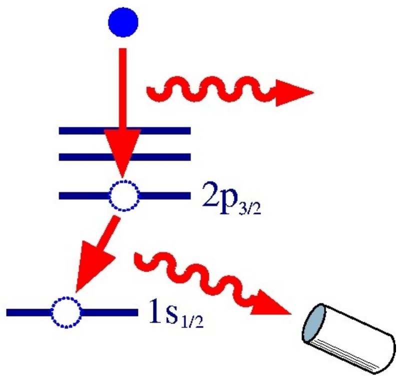


S. Fritzsche et al., PRL 103 (2009) 113001.

Z. Hu et al., PRL 108 (2012) 073002 (exp. confirmation).

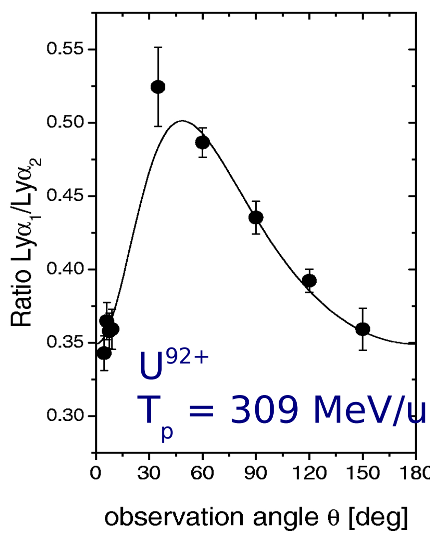
Details matter:

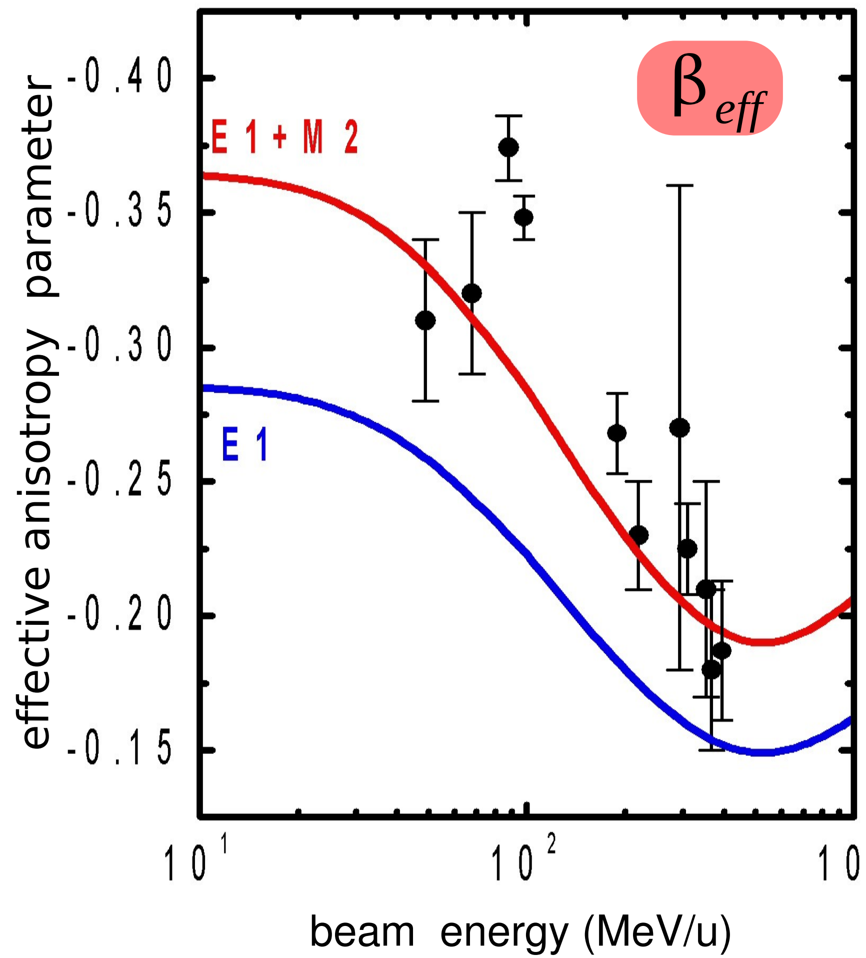
-- Lyman-a vs. K-a emission from high-Z ions



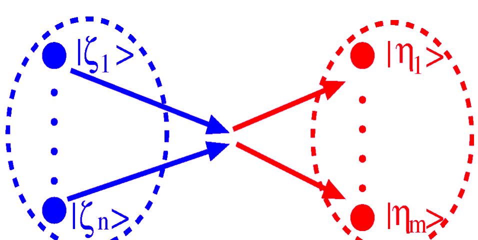
(initially) bare ion

Ly-α_1 is strongly anisotropic





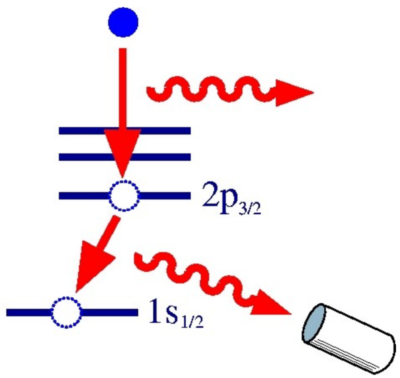
Initial state $(t \to -\infty)$ $\hat{\rho}_i$



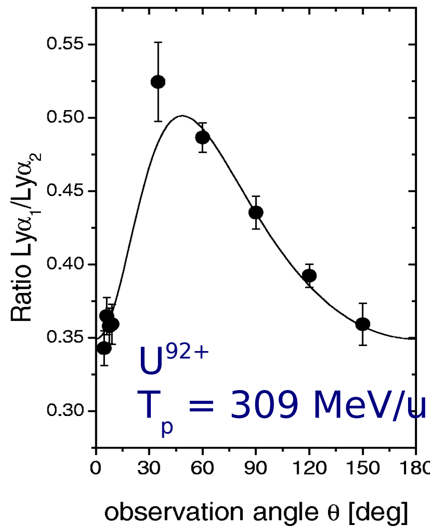
Final state $(t \to +\infty)$ $\hat{\rho}_f$

Details matter: Adding one electron

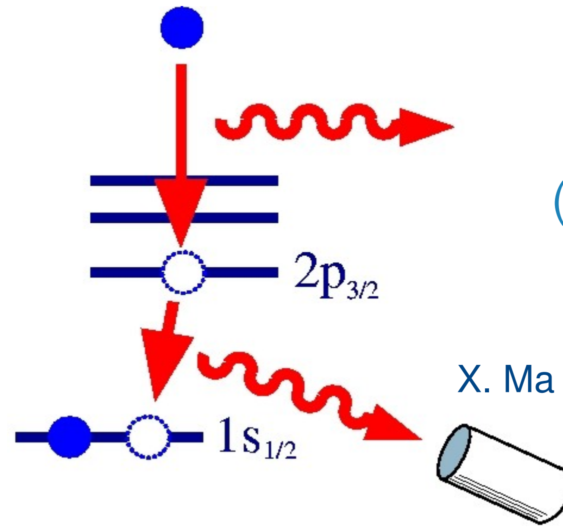
-- Lyman-a vs. K-a emission from high-Z ions



(initially) bare ion

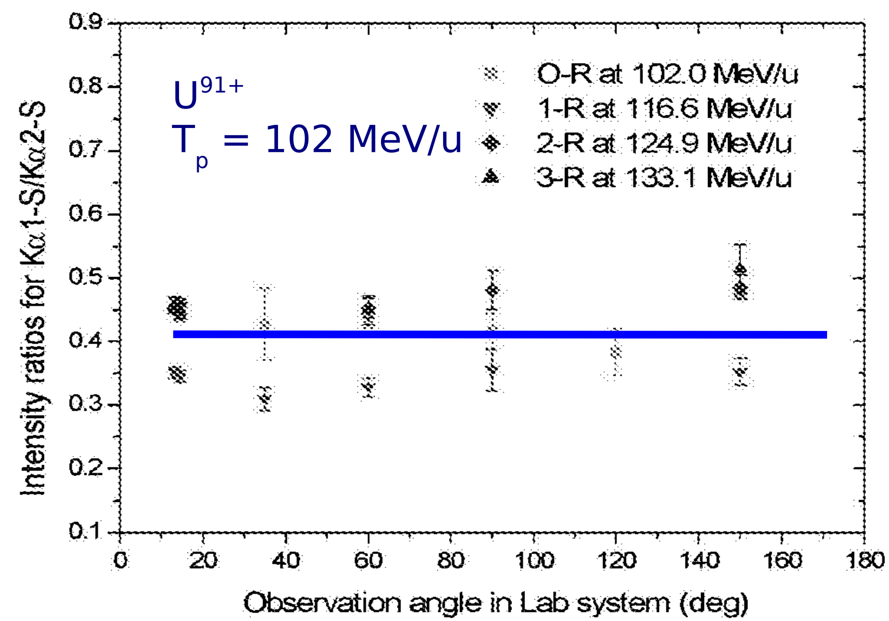


Ly-α_1 is strongly anisotropic



(initially) H-like ion

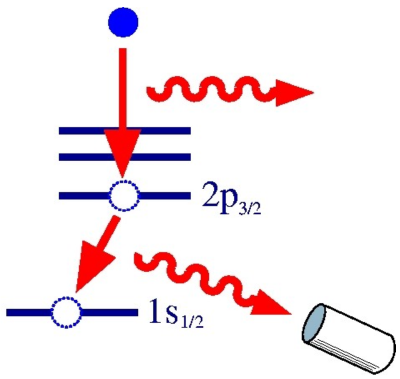
X. Ma et al, PRA 68 (2003) 042712.



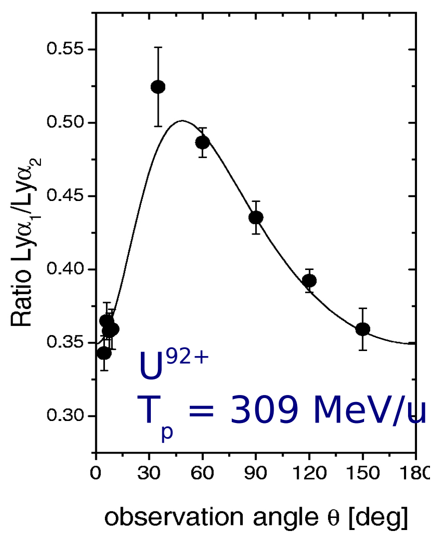
K-α_1 is isotropic

Details matter: Adding one electron

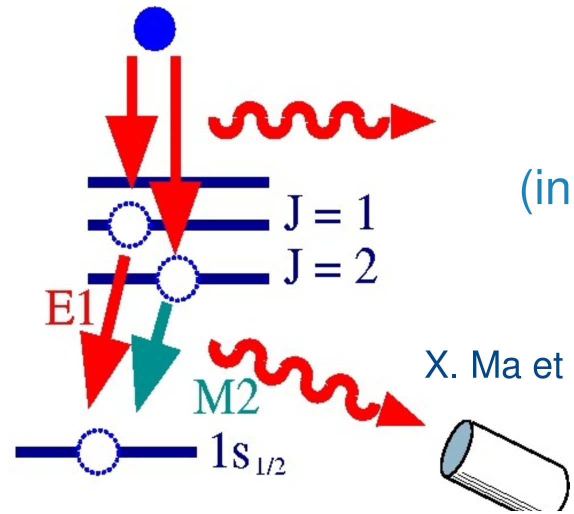
-- Lyman-α vs. K-α emission from high-Z ions



(initially) bare ion

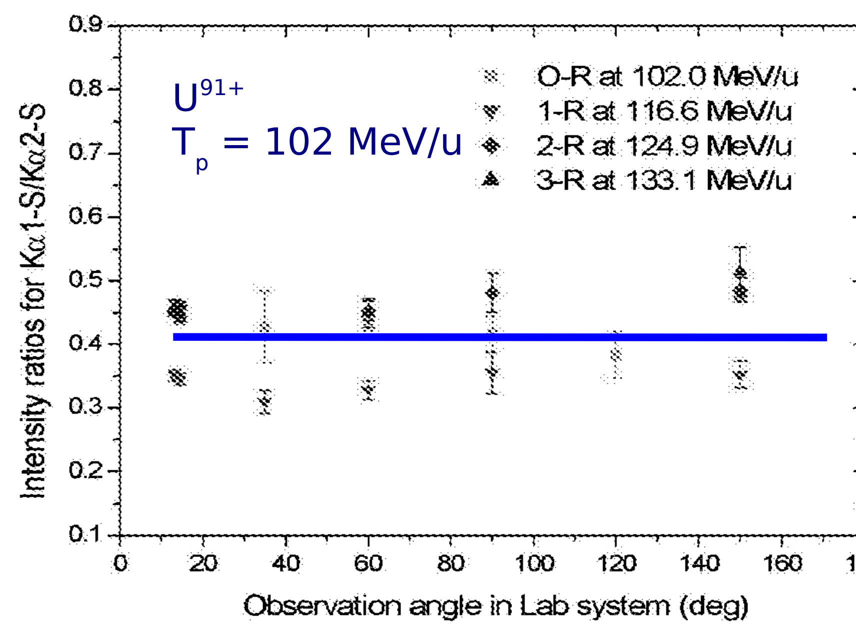


Ly-α_1 is strongly anisotropic



(initially) H-like ion

X. Ma et al, PRA 68 (2003) 042712.



K-α_1 is isotropic

E1: $$W(\theta)_{E1} \sim 1 + \frac{1}{\sqrt{2}} A_2(J=1) P_2(\cos\theta)$$

M2: $$W(\theta)_{M2} \sim 1 - \sqrt{\frac{5}{14}} A_2(J=2) P_2(\cos\theta)$$

K-α decay of highly-charged ions

-- angular distribution as „observed“ in experiments

$$W(\theta)_{K\alpha_1} \sim N_{J=1} W_{E1}(\theta) + N_{J=2} W_{M2}(\theta)$$

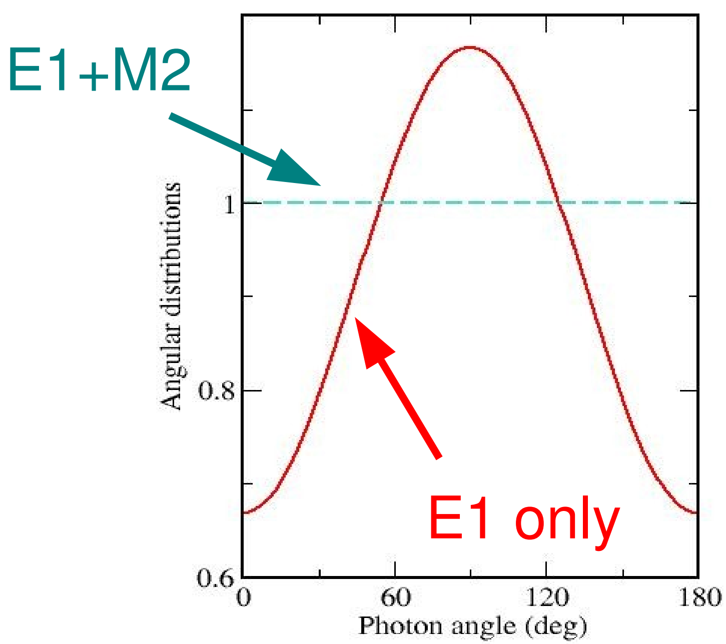
A. Surzhykov et al., PRA 73 (2006) 032716.

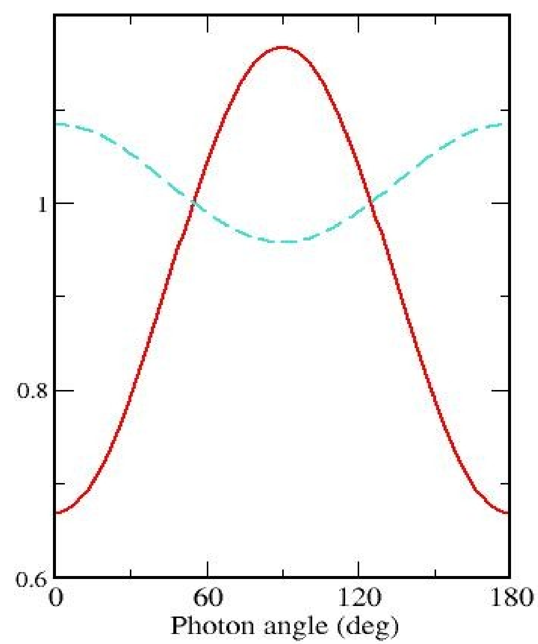
$$= 1 + \left(N_{J=1} \frac{1}{\sqrt{2}} A_2(J=1) - N_{J=2} \sqrt{\frac{5}{14}} A_2(J=2) \right) P_2(\cos\theta)$$

$N_{J=1}, N_{J=2}$ relative populations of J=1, 2 states

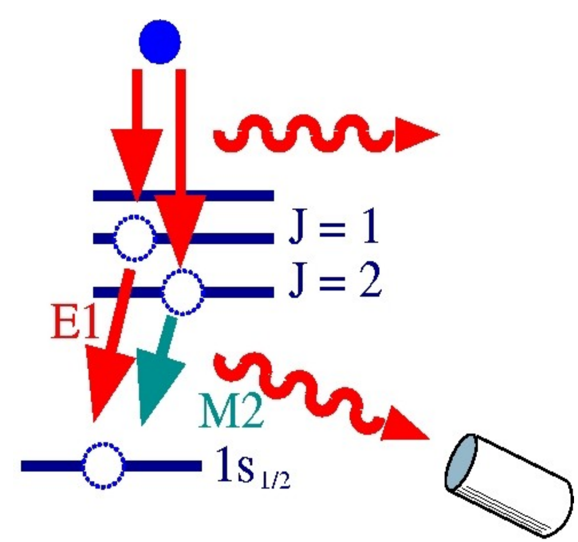
$$N_{J=1} = N_{J=2} = \frac{1}{2}$$

$$N_{J=1} = \frac{3}{8} \quad N_{J=2} = \frac{5}{8}$$





Calculations have been done for L-REC of U^{91+} with T_p = 100 MeV/u



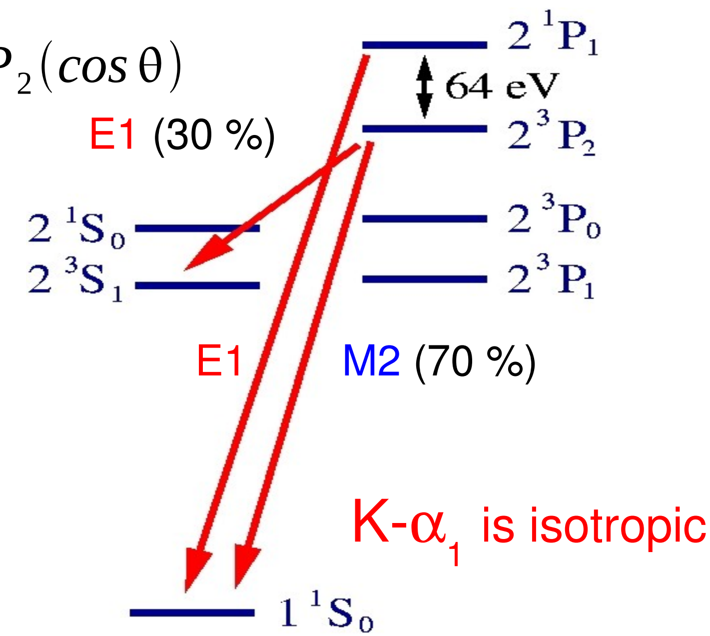
K-α decay of highly-charged ions

-- for 220 MeV/u U^{90+} ions following REC

$$W(\theta)_{K\alpha_1} \sim N_{J=1} W_{E1}(\theta) + N_{J=2} W_{M2}(\theta)$$

A. Surzhykov et al., PRA 73 (2006) 032716.

$$= 1 + \left(N_{J=1} \frac{1}{\sqrt{2}} A_2(J=1) - N_{J=2} \sqrt{\frac{5}{14}} A_2(J=2) \right) P_2(\cos\theta)$$



- Relative populations of the J = 1, 2 levels following REC (IPM model):

$$\frac{N_{J=1}}{N_{J=2}} = \frac{3}{5}$$

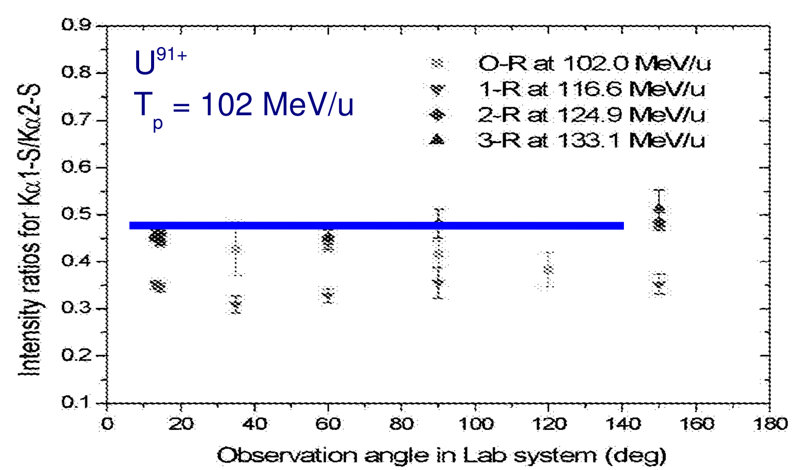
- By taking into account $^3P_2 \to {}^3S_1$ channel:

$$\frac{N_{J=1}}{N_{J=2}} = \frac{6}{7}$$

initial `capture' populations

+ branching fractions

$$\boxed{\left(\frac{N_{J=1} - N_{J=2}}{N_{J=1} + N_{J=2}} \right)_{theory} \approx -0.08}$$



K-α decay of highly-charged ions

-- following the Coulomb excitation of the projectiles

$$W(\theta)_{K\alpha_1} \sim N_{J=1} W_{E1}(\theta) + N_{J=2} W_{M2}(\theta)$$

$$= 1 + \left(N_{J=1} \frac{1}{\sqrt{2}} A_2(J=1) - N_{J=2} \sqrt{\frac{5}{14}} A_2(J=2) \right) P_2(\cos\theta)$$

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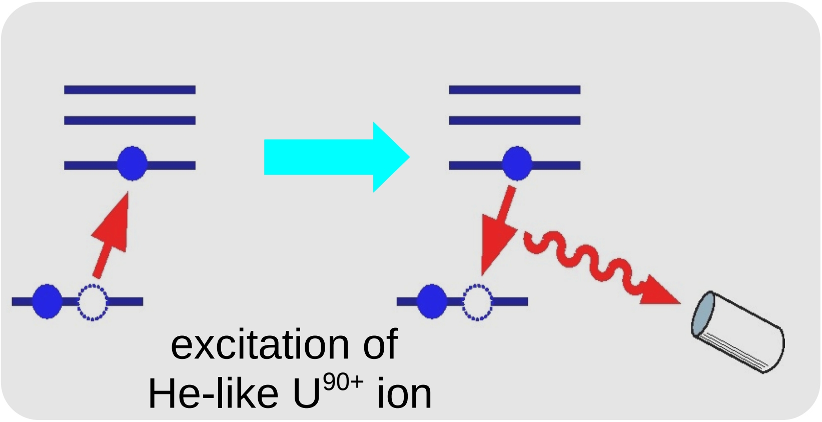
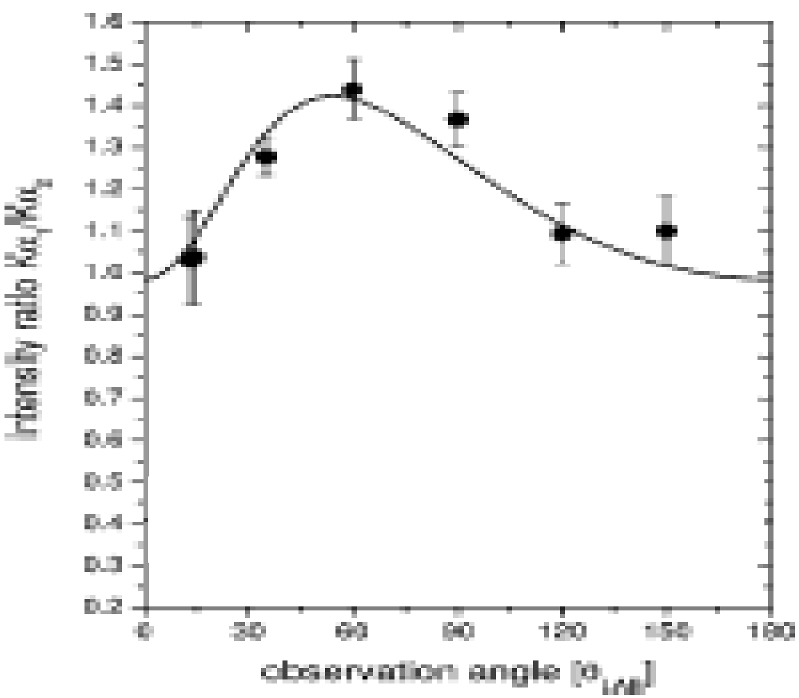




- Excited states of He-like heavy ions can be produced also by the Coulomb excitation of the projectile in the field of target atoms.
- Experiments were already performed at the GSI storage ring for He-like uranium ions U^{90+}.
- Strong anisotropy of the subsequent Ka_1 radiation has been observed!

K-α decay of highly-charged ions

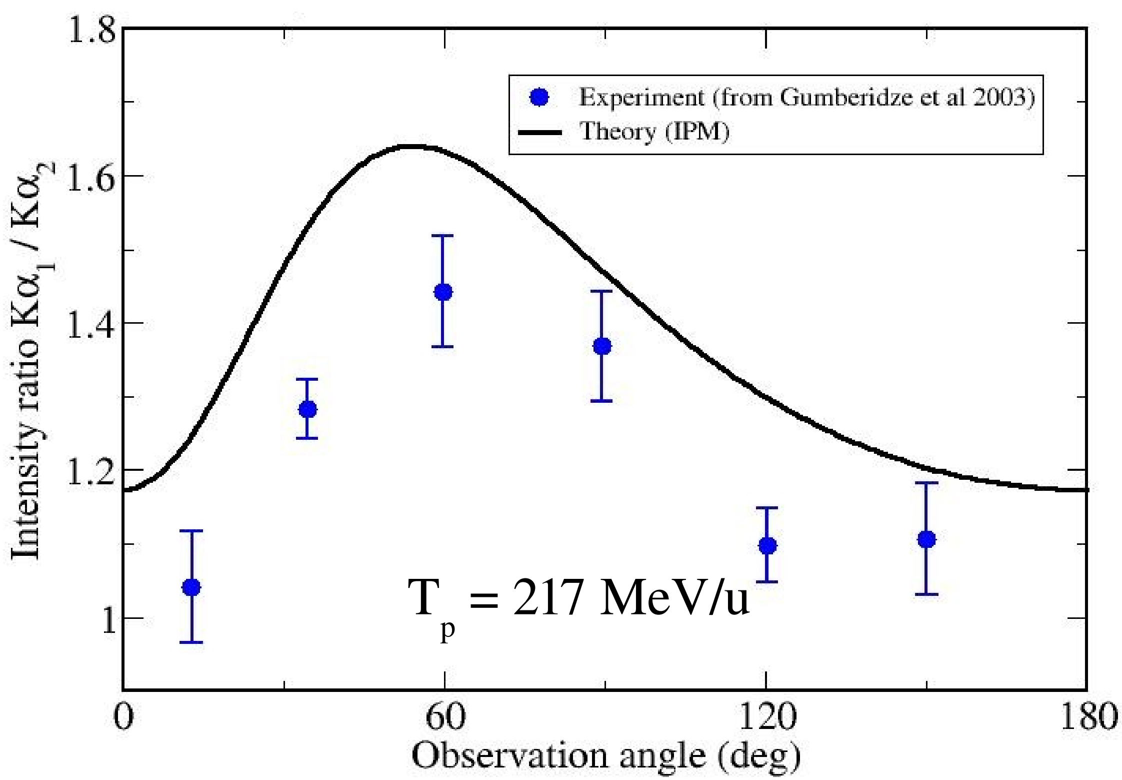
-- following the Coulomb excitation of the projectiles

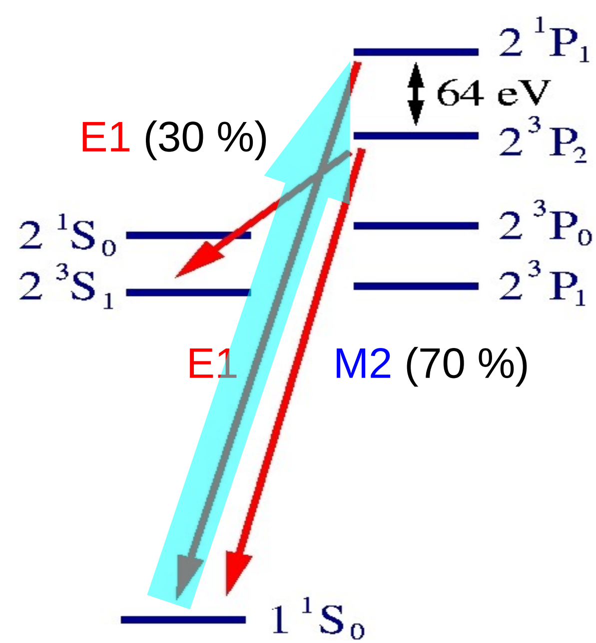
$$W(\theta)_{K\alpha_1} \sim N_{J=1} W_{E1}(\theta) + N_{J=2} W_{M2}(\theta)$$

$$= 1 + \left(N_{J=1} \frac{1}{\sqrt{2}} A_2(J=1) - N_{J=2} \sqrt{\frac{5}{14}} \cancel{A_2(J=2)}\right) P_2(\cos\theta)$$

$N_{J=1}, N_{J=2}$ relative populations of J=1, 2 states

- Angular distribution results dominantly from the decay of the J=1 level.
- Role of electron-electron interactions still unexplored.





Techniques and tools for strong-field processes

– to describe the structure & dynamics 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- and few-electron atoms in such a way to enable the „computation“ of complex systems and processes.

Techniques and tools for strong-field processes

– to describe the structure & dynamics of atoms and ions

<u>Multiconfiguration expansions</u>

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

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

Many-particle character
„electronic correlations“

<u>Energies & Wave functions</u>

- Cowan / CIV3
- MCHF
- GRASP(-92) / RATIP
- „Desclaux“
- Coupled-Cluster

Shell structure
static vs. dynamic correlations

Generalization of the knowledge a
in such a way to enable the „comp

Techniques and tools for strong-field processe

– to describe the structure & dynami ns

Multiconfiguration expansions

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

Construct a `physically motivated'
in the N-electron Hilbert spac

There is no simple path from one element to the next,
from one ionization process to the other,
from one property to ...

Many

relations

Generalization of the knowledge a
in such a way to enable the „comp

Energies & Wave functions

- Cowan / CIV3
- MCHF
- GRASP(-92) / RATIP
- „Desclaux“
- Coupled-Cluster

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.

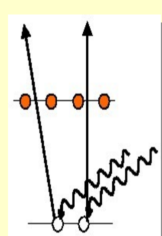
REOS: Relaxed-orbital Einstein A and B coefficients, transition probabilities and lifetimes.

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

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

Many-electron basis (wave function expansions)

- Construction and classification of N-particle Hilbert spaces
- Shell model: Systematically enlarged CSF basis
- Interactions
  - Dirac-Coulomb Hamiltonian
  - Breit interactions + QED
  - Electron continuum; scattering phases
- Coherence transfer and Rydberg dynamics

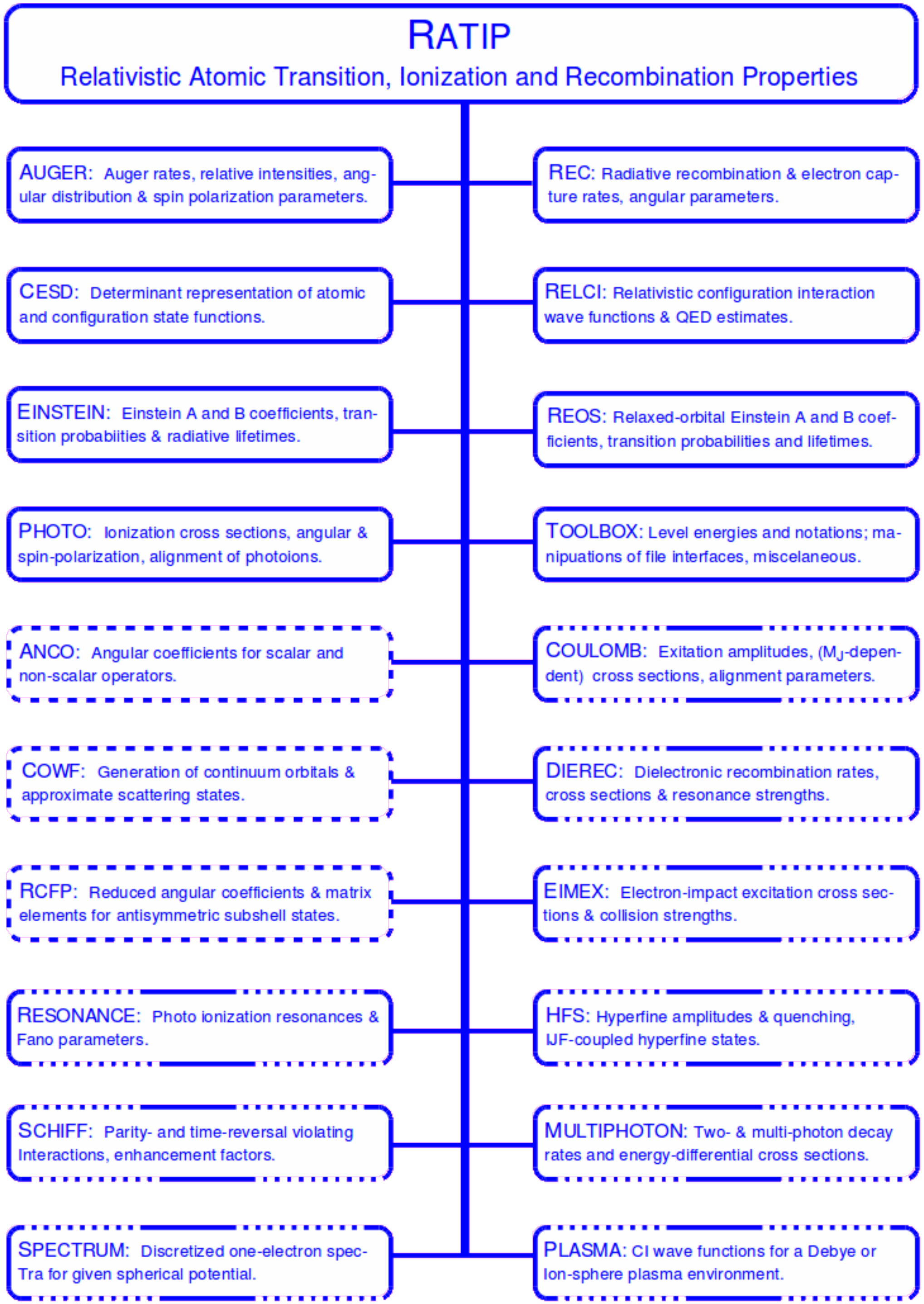


S. Fritzsche, JESRP 114-116 (2001) 1155; Comput. Phys. Commun. 183 (2012) 1525

Multiconfiguration expansions

$$\psi_\alpha(PJM) = \sum_r^{n_c} c_r(\alpha)\,\gamma_r PJM>$$

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



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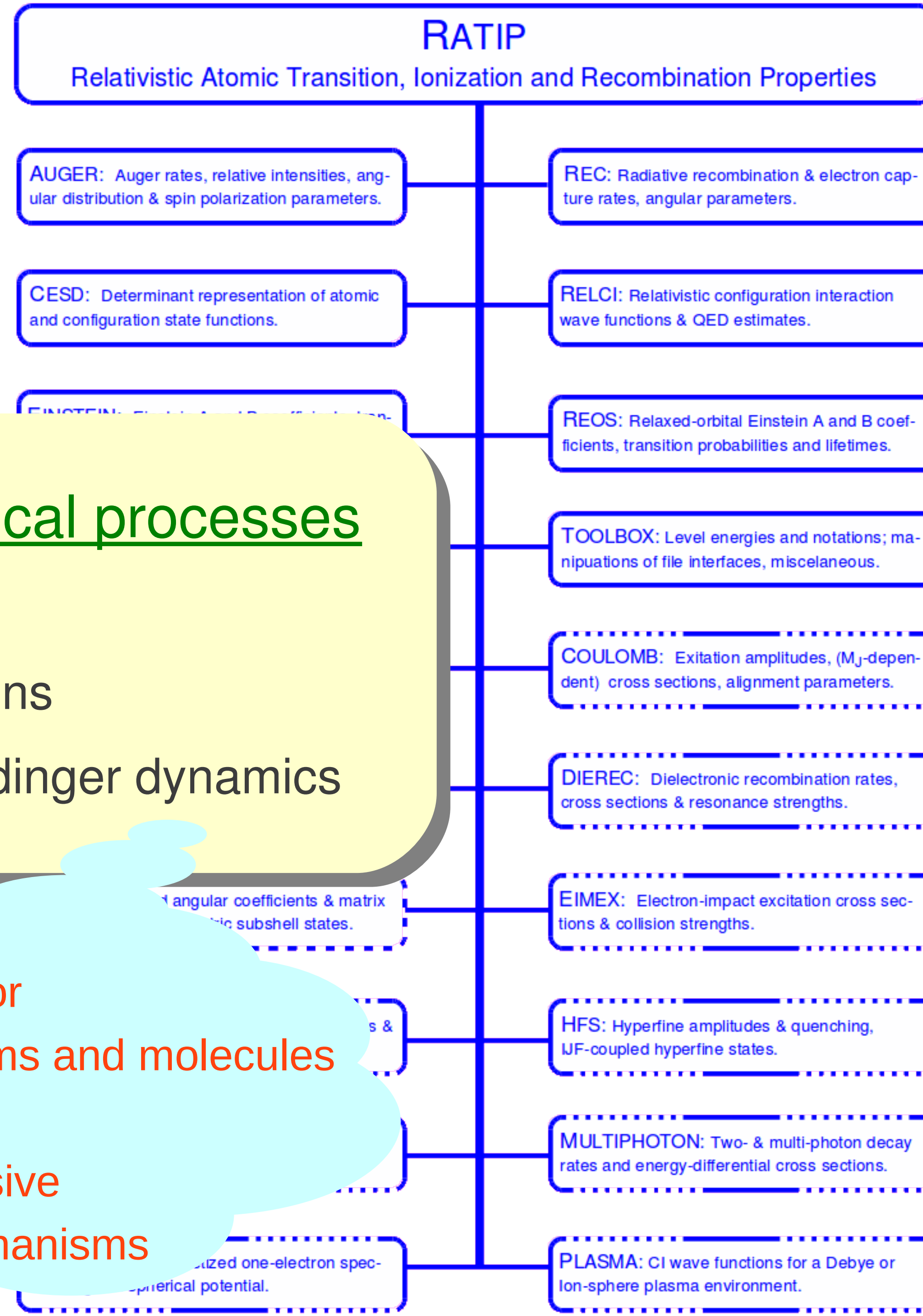


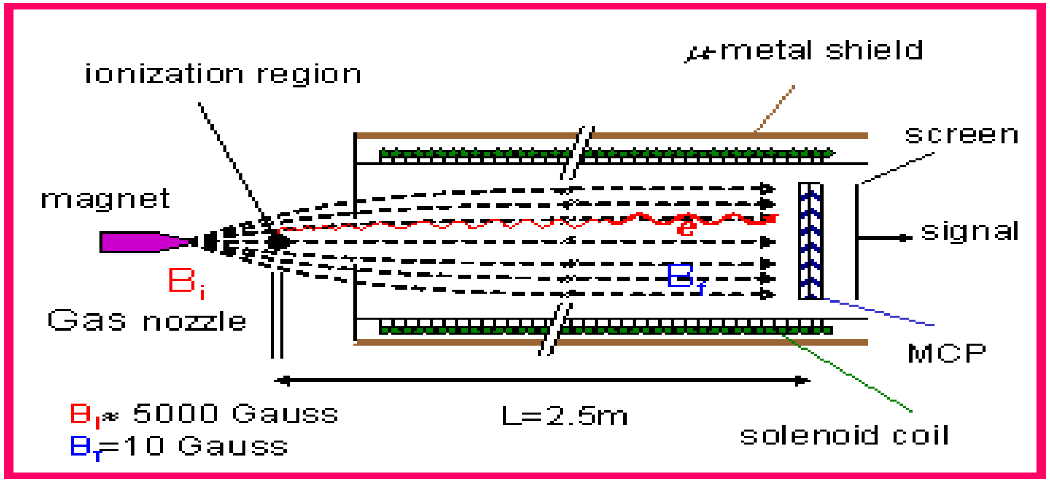
Frameworks for studying dynamical processes

- perturbative vs. non-perturbative
- rate equations vs. electronic correlations
- (in-) coherent evolution vs. full Schrödinger dynamics

- Impractical (unfeasible) for most atoms and molecules
- No physical insight
- (numerically) very expensive
- Little control about phys. mechanisms

Multiple ionization of noble gases

-- using magnetic bottle techniques



P. Kruit and F. H. Read, J. Phys. E <u>16</u>, 313 (1983)
J. H. D. Eland et al, Phys. Rev. Lett. <u>90</u>, 053003 (2003)

Advantages

- Electron detection with high sensitivity and resolution (especially charge-particle detect.)
- Supports both, normal TOF but also zero-kinetic energy (resolution)
- High collection efficiency (reconstruction of 3D velocity distributions)
- Coincidence maps: Differential analysis of outgoing electrons
- Easy to combine with atomic and molecular beams
- ☆ No information about angular distribution and spins of photoelectrons

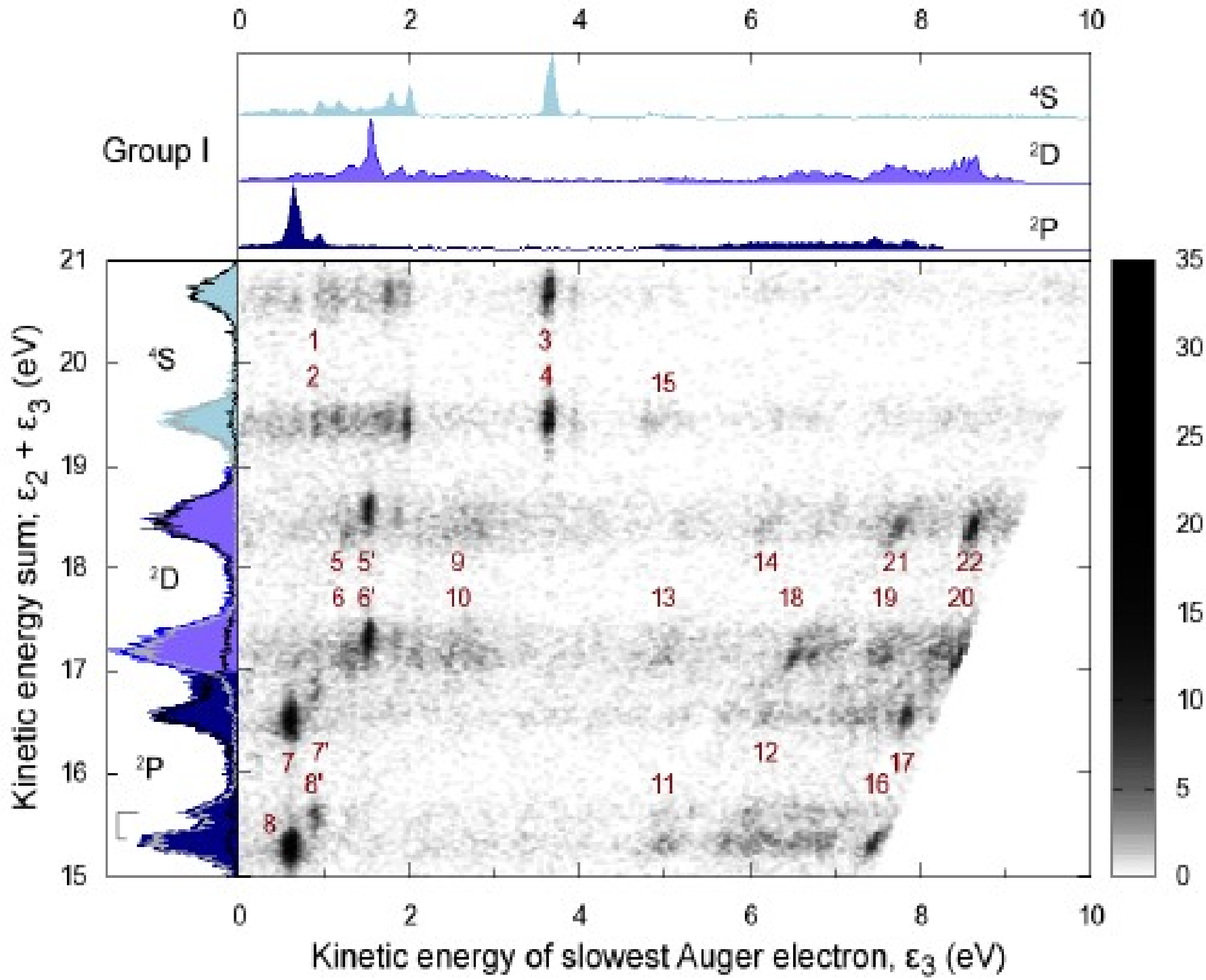
Multiple ionization of noble gases @ synchrotrons

-- 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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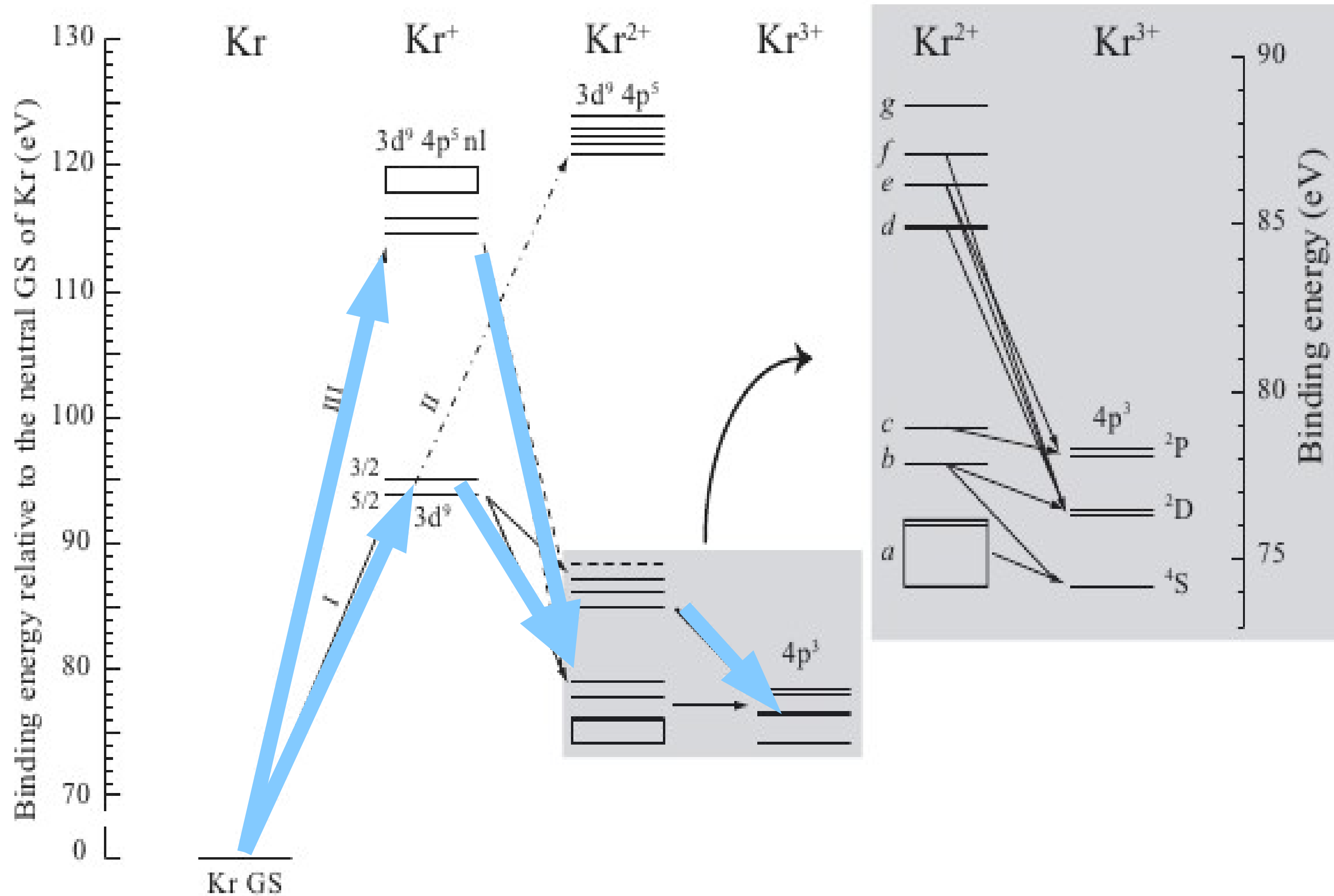

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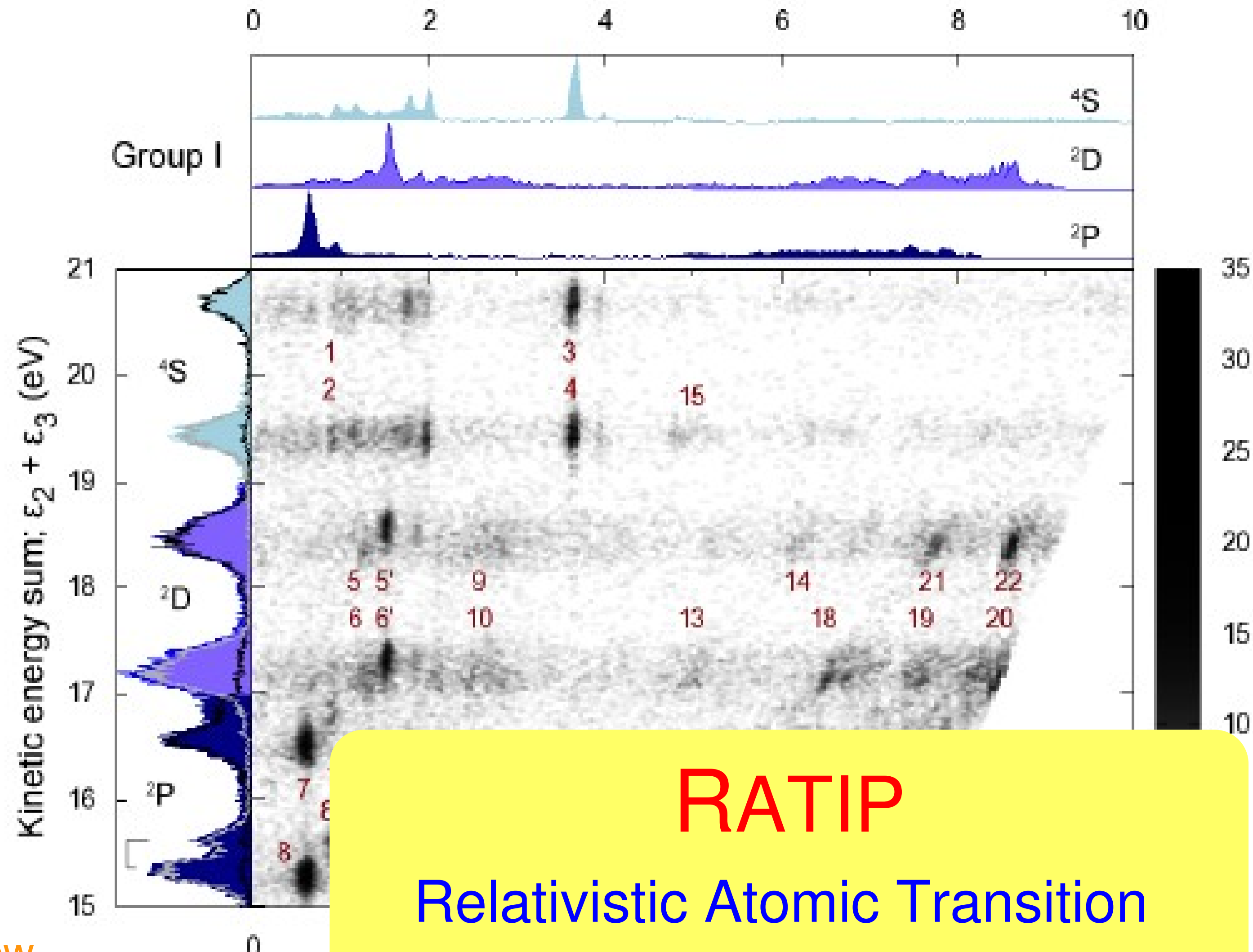


Calculations helped identify new levels and decay pathes in the sequential (auto-) ionization.

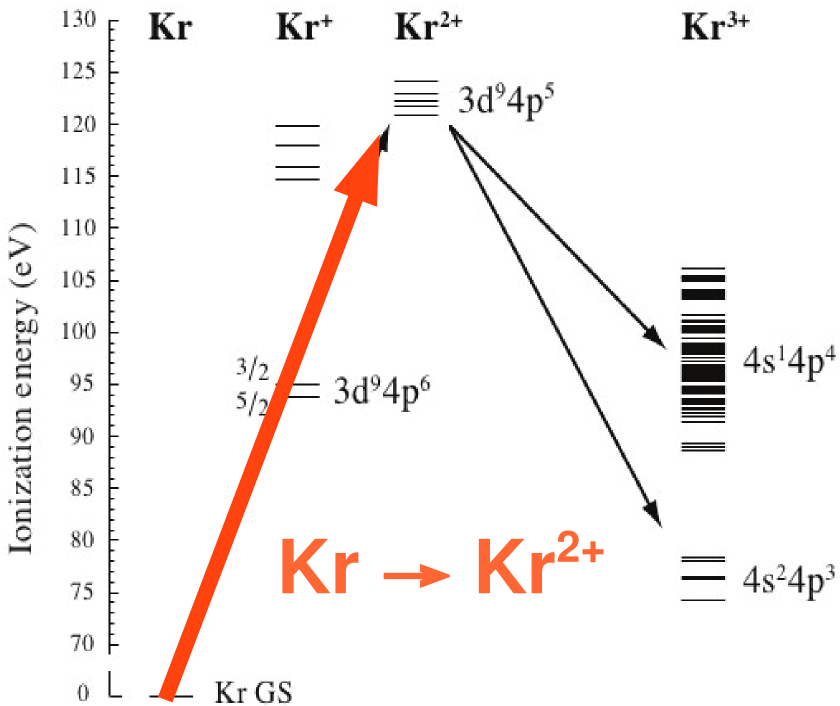
RATIP

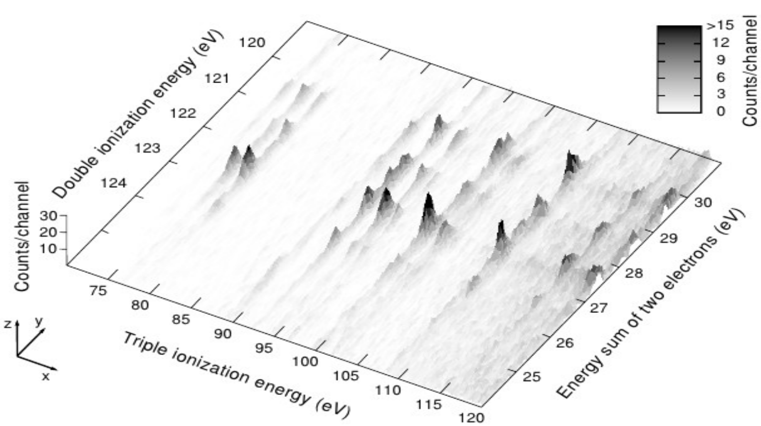
Relativistic Atomic Transition and Ionization Properties

(CPC library)

Formation of Kr^{3+} ions

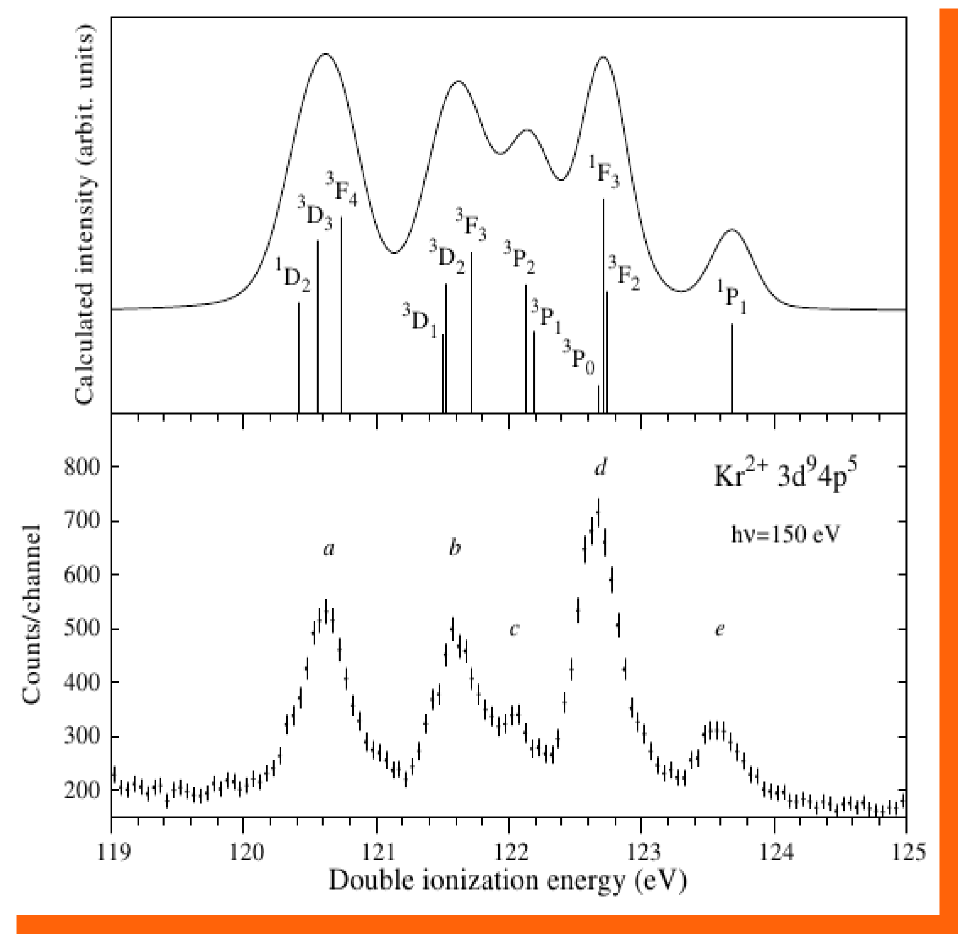
– via core-valence doubly ionised intermediate states





Single photon core-valence double ionisation

Spectra of krypton recorded at hν= 150 eV.



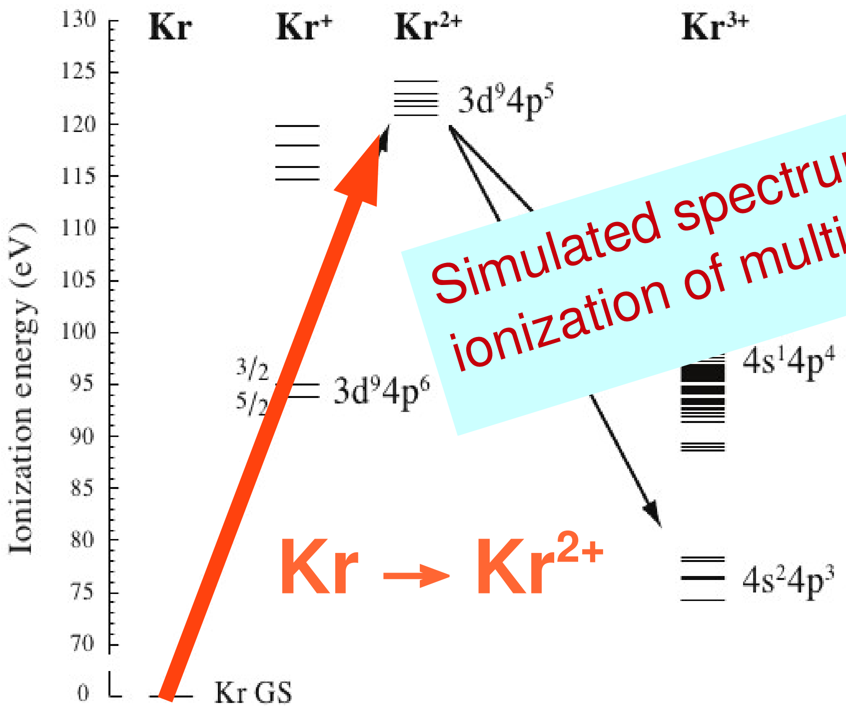
E. Andersson et al, PRA 82 (2010) 043418;
PRA 85 (2012) 032502.

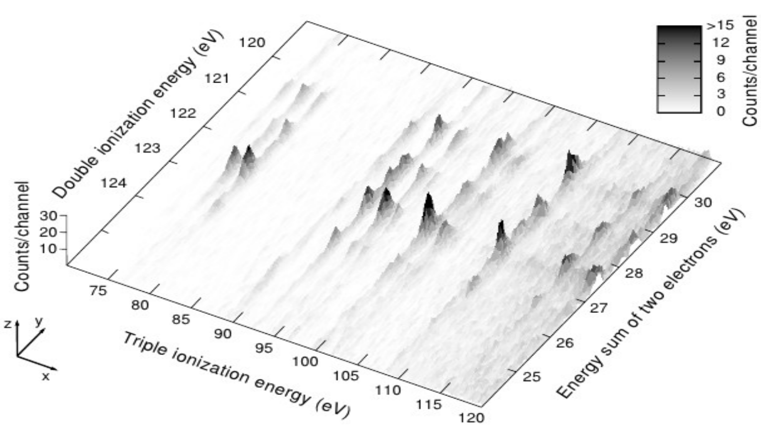
Formation of Kr^{3+} ions

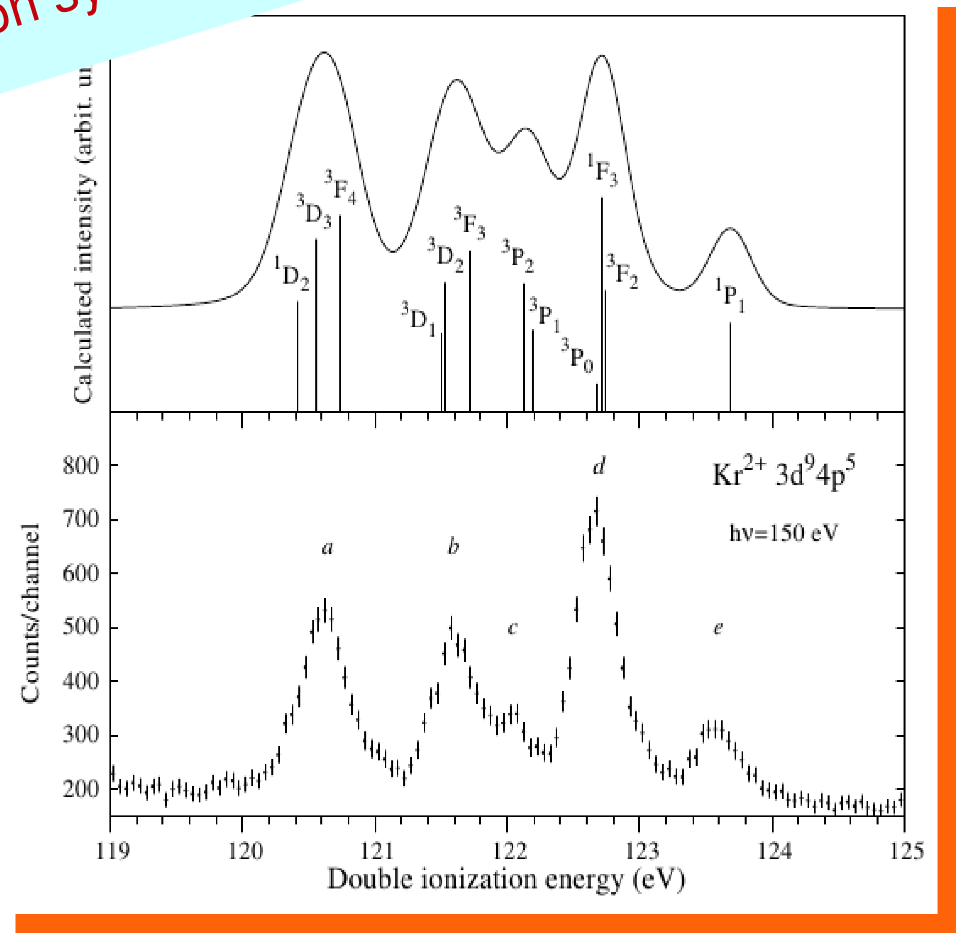
– via core-valence doubly ionised intermediate states

Simulated spectrum for direct double ionization of multi-electron systems !!

Si... core-valence double ionisation
...oton recorded at hv= 150 eV.







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Formation of Kr^{3+} ions

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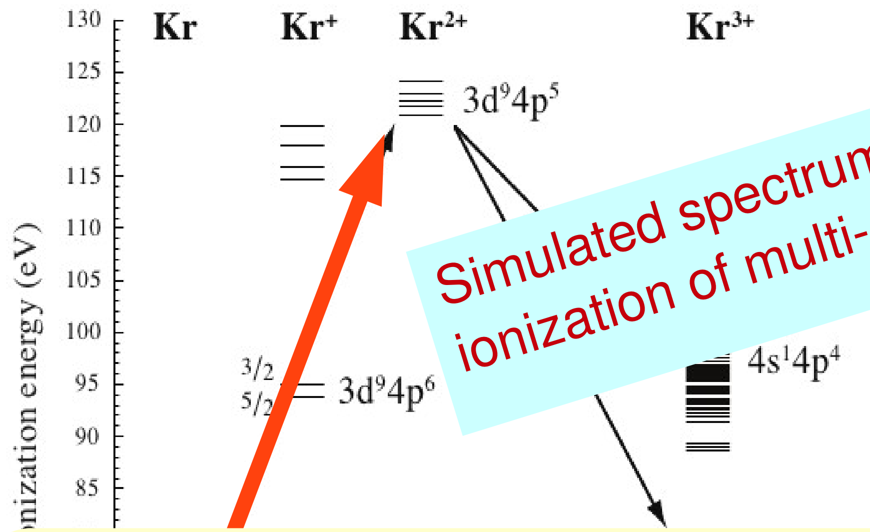
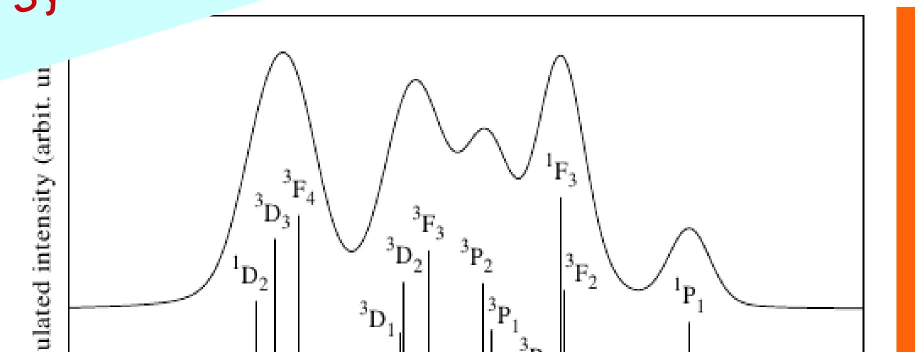


Simulated spectrum for direct double ionization of multi-electron systems !!

... core-valence double ionisation

...oton recorded at hv= 150 eV.

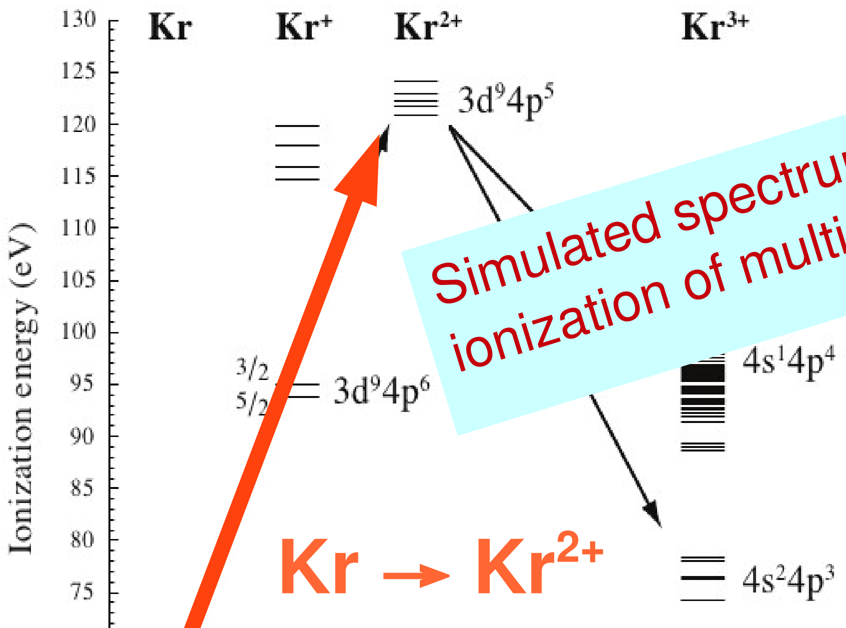


Features of the *ab-initio* approach

- Combines time-independent density matrix with proper construction of `two-electron' ionization amplitudes.
- Coupling of wave packets from discretization of the one-electron spectrum.
- Two-electron continuum through direct diagonalization (damped CI vs. finite e-e interaction with r_{cut}).
- Explicit summation over intermediate many-electron states (continua).

Formation of Kr^{3+} ions

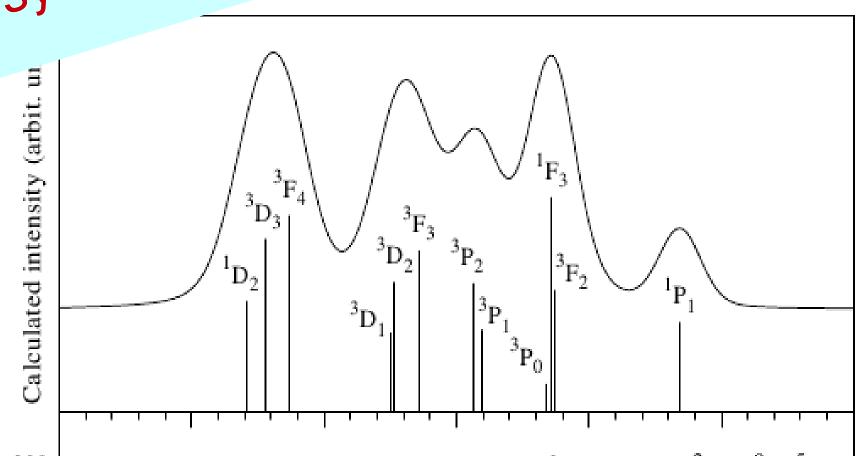
– via core-valence doubly ionised intermediate states



$Kr \rightarrow Kr^{2+}$

Si… core-valence double ionisation
…oton recorded at hv= 150 eV.

Simulated spectrum for direct double ionization of multi-electron systems !!



Approach works well:

+ if DI is insensitive with regard to energy sharing
+ core-core and core-valence ionization (localized electron density)
+ reasonably high photon energies

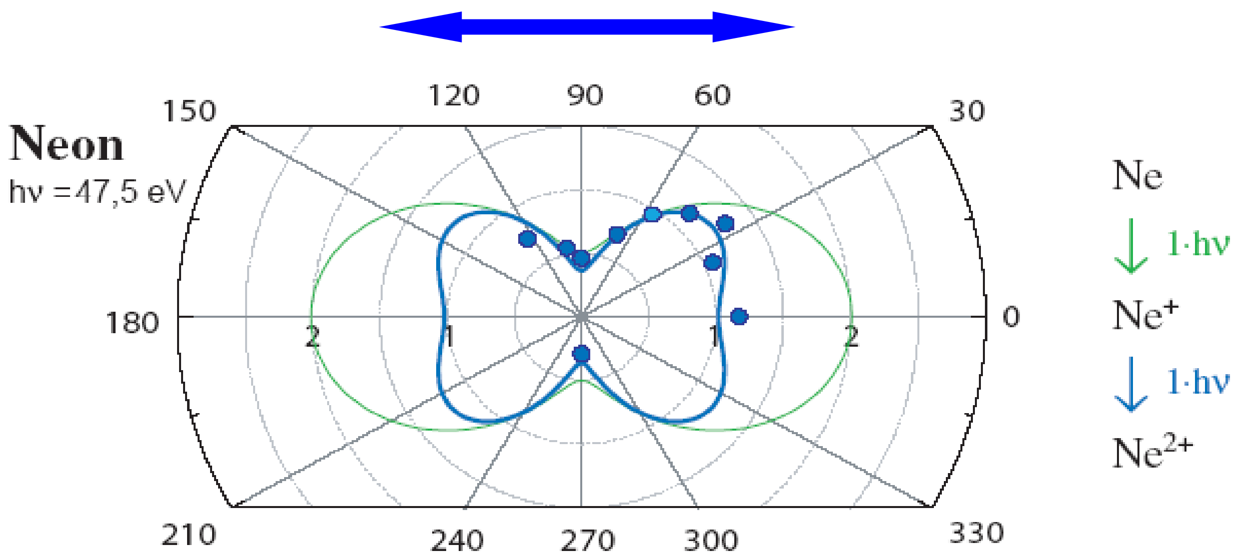
– more details desirable about energy sharing and angular dependence

– more multi-electron spectra, other elements and shell structures.

Light-matter interactions at increased (FEL) intensities

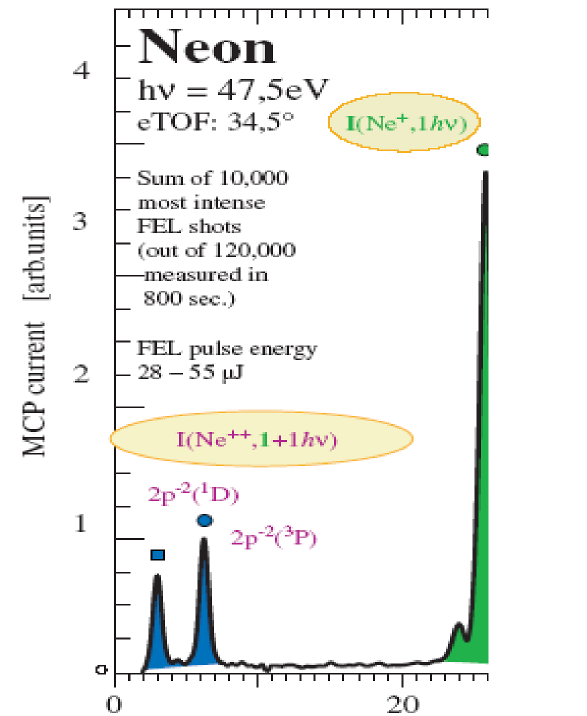
– first non-linear studies on electron emission

Details in the ionization dynamics of electrons became visible first in the sequential and direct DI of nobles gases



$2p^{-1}$: $I(\theta) = C\left[1 + \beta_2 P_2(\cos\theta)\right]$ $\beta_2 = 1.00$

$2p^{-2}(^3P)$: $I(\theta) = C\left[1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta)\right]$ $\beta_2 = 0.74,$ $\beta_4 = -0.50$



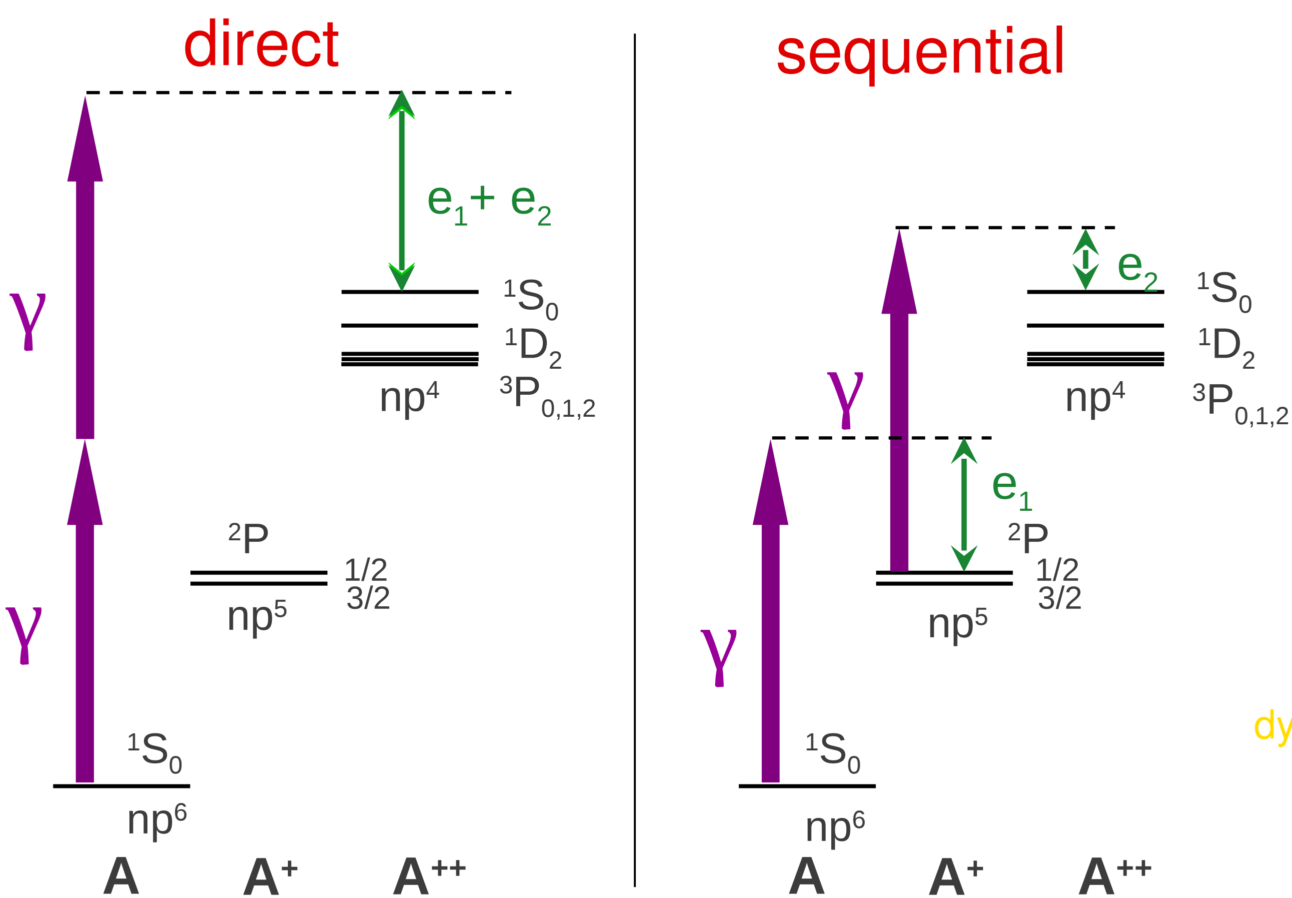
M. Braune et al, ICPEAC (2007/2009).

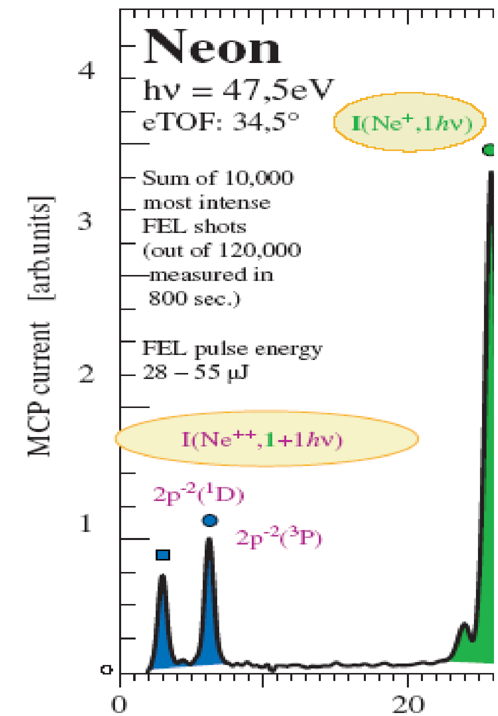
Collaboration with A. Grum-Grzhimailo, E. Gryzlova & N. Kabachnik

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).

$$\frac{dN^{+}}{dt} = \frac{N}{T}\,\sigma^{(0\to1)}\left(\frac{\mathcal{N}_{\rm ph}}{A}\right)$$

dynamical coupling ?

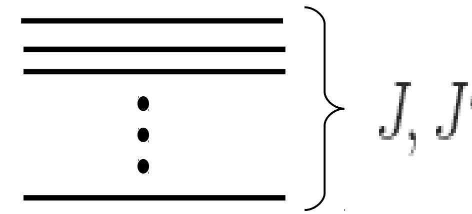
$$\frac{dN^{2+}}{dt} = \frac{N}{T}\,\sigma^{(0\to2)}\left(\frac{\mathcal{N}_{\rm ph}}{A}\right)^{2}$$

Depolarization due to multiple (auto-) ionization

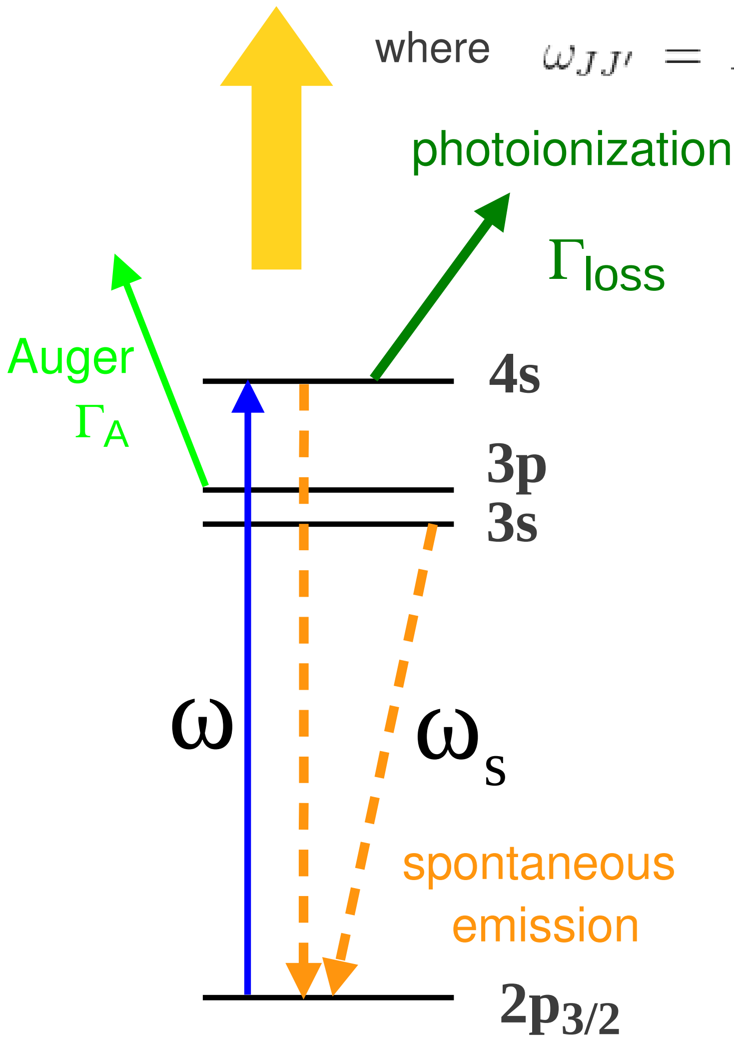
– exotic matter states with multiple inner-shell holes

Quantum evolution of the density operator:

$$\rho_{kq}(J,J';t) = \rho_{kq}(J,J';t_1)\exp[(i\omega_{JJ'} - \Gamma_{JJ'})(t-t_1)]$$



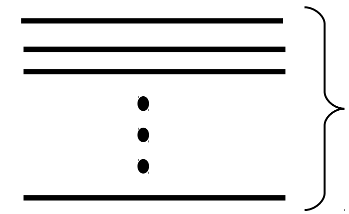
where $\omega_{JJ'} = E_J - E_{J'} \quad \Gamma_{JJ'} = (\Gamma_J + \Gamma_{J'})/2$



Depolarization due to multiple (auto-) ionization

– interplay of different time scales leads to depolarization

Quantum evolution of the density operator:

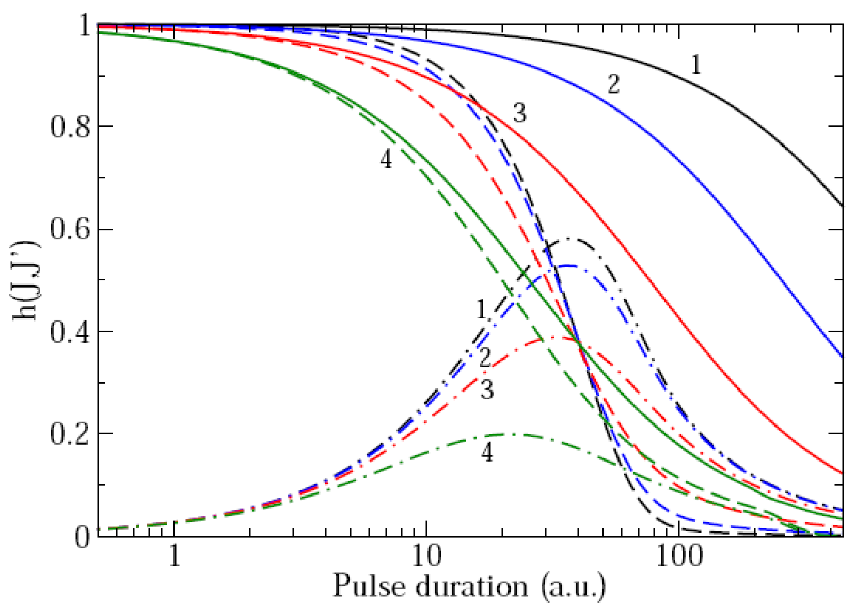


$$\rho_{kq}(J, J'; t) = \rho_{kq}(J, J'; t_1) \exp[(i\omega_{JJ'} - \Gamma_{JJ'})(t - t_1)]$$

where $\omega_{JJ'} = E_J - E_{J'} \quad \Gamma_{JJ'} = (\Gamma_J + \Gamma_{J'})/2$

Depolarization factor in the e-e correlation function

$$h(J, J') = \int_{-\infty}^{+\infty} dt_1\, I(t_1) \int_{t_1}^{\infty} dt_2 \exp[(i\omega_{JJ'} - \Gamma_{JJ'})(t_2 - t_1)]\, I(t_2)$$



Two coherently excited levels with

$w_{JJ'} = 0.025$ a.u.

solid: diagonal terms J = J'

dashed: nondiagonal terms

1: $\Gamma = 0.001$ a.u.

2: $\Gamma = 0.003$ a.u.

3: $\Gamma = 0.01$ a.u.

4: $\Gamma = 0.03$ a.u.

Coupling to the electron continuum

S. Fritzsche et al., JPB 44 (2011) 175602.

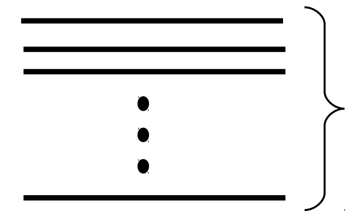
"JPB Highlight 2011"

Depolarization due to multiple (auto-) ionization

– non-linear response to intense ultra-short x-rays

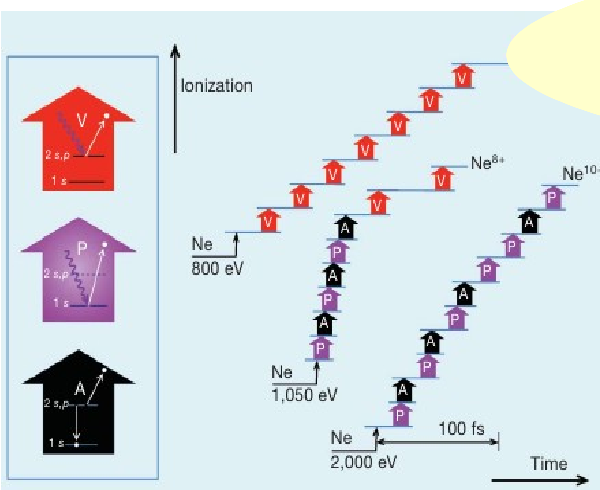
Quantum evolution of the density operator:

$$\rho_{kq}(J, J'; t) = \rho_{kq}(J, J'; t_1)\exp[(i\omega_{JJ'} - \Gamma_{JJ'})(t - t_1)]$$

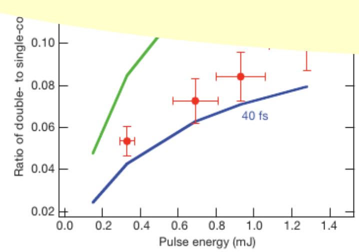


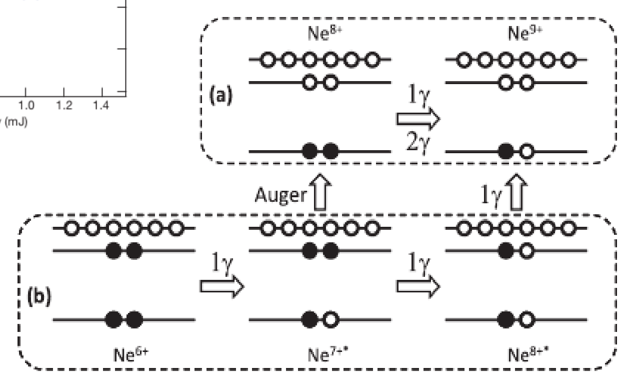
where $\omega_{JJ'} = E_J - E_{J'} \quad \Gamma_{JJ'} = (\Gamma_J + \Gamma_{J'})/2$

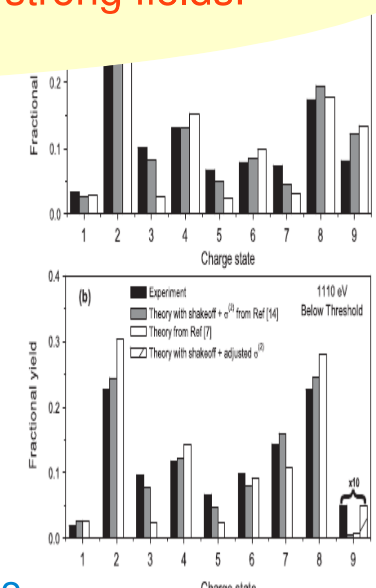
To proceed beyond `rate-equation' dynamics in understanding the femto-second electronic response of atoms in strong fields.



L. Young et al., Nature 466 (2010) 56.





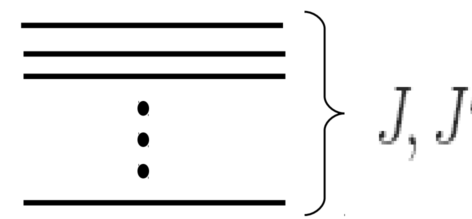


G. Doumy et al., PRL 106 (2011) 083002.

Depolarization due to multiple (auto-) ionization

– non-linear response to intense ultra-short x-rays

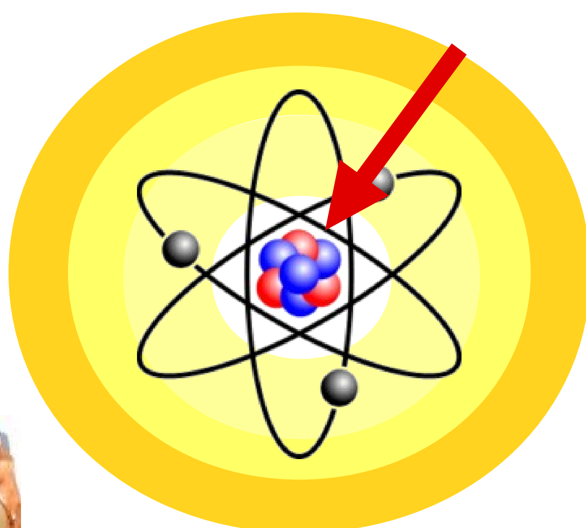
Quantum evolution of the density operator:



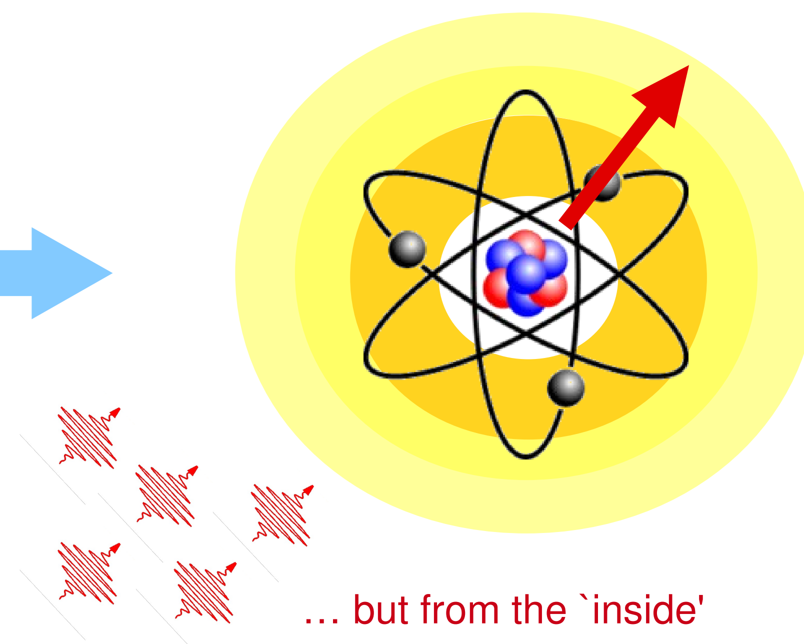
$$\rho_{kq}(J, J'; t) = \rho_{kq}(J, J'; t_1) \exp[(i\omega_{JJ'} - \Gamma_{JJ'})(t - t_1)]$$

where $\omega_{JJ'} = E_J - E_{J'}$ $\Gamma_{JJ'} = (\Gamma_J + \Gamma_{J'})/2$

Intense optical and VUV laser



intense FEL radiation



peels off the electrons layer by layer

... but from the `inside'

Coherent time evolution of inner-shell excited systems

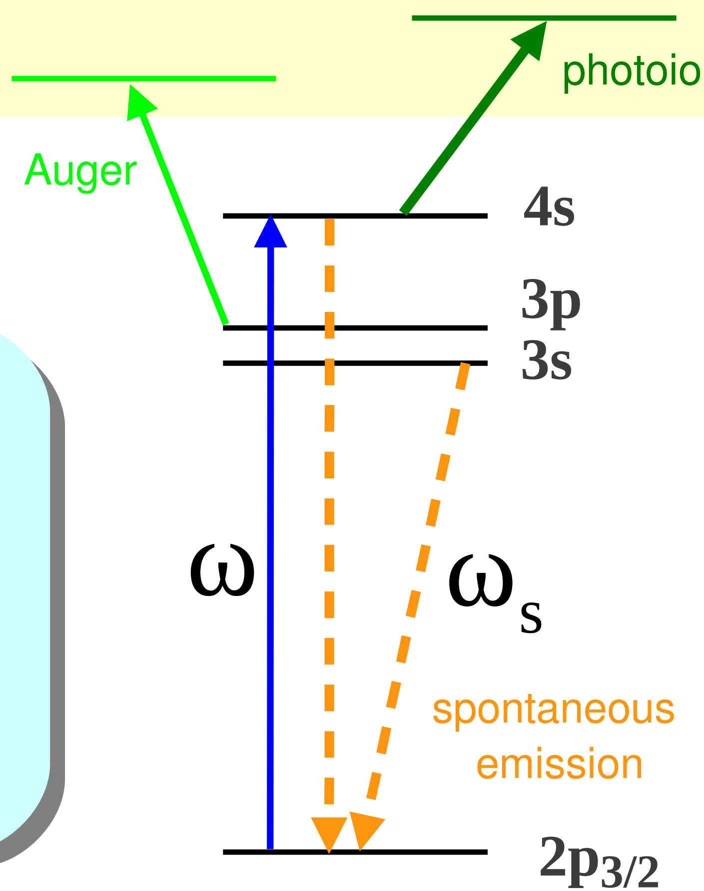
– in short-pulse or pump-probe experiments

Explicitly time-dependent density operator:

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

Based on Liouville's equation

Including (spatial) dynamics
of emitted electrons & photons



Frequently applied in quantum optics:

- N (non-degenerate) bound levels
- long pulses with linear polarization
- electric-dipole & RWA approximation
- influence of coupling fields

(EIT, slow light, ...)

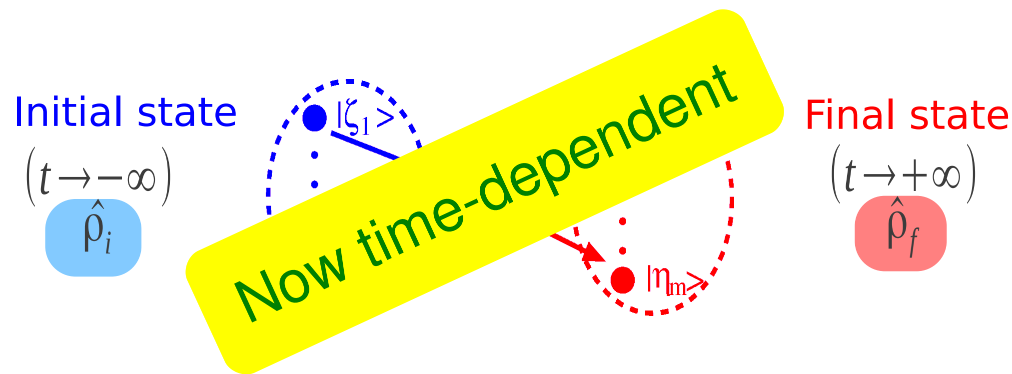
Coherent time evolution of inner-shell excited systems

– 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



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

direct coupling

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

exchange

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

ionization & loss processes

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

S. Fritzsche, CPC 183 (2012) 1525

Collaboration with Alexei Grum-Grzhimailo

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:

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

Now time-dependent

Based Liouville's equation

$i\,\frac{d}{}$

Atomic (

amplitu

S. Fritzsch

Collaboratio

The "hope": Compared with time-dependent SE

- control of approximations: RWA, dipole, … based on "physical insight"
- explicit electron dynamics

  (including angular-and spin-dependent emission)
- pulses with given shape and polarization
- higher multipole contributions
- (rather) simple control upon effects of level and pulse structures
- feasibe for many-electron systems in 3D

Summary

- Intense FEL pulses lead to „hollow atoms“ and ions, i.e. extreme matter in a truly exotic form. Dynamics of strongly correlated electrons in controlled light fields is a presently emerging field and require the coherent time evolution of the systems.
- For atoms & molecules, multiphoton ionization and autoionization is likely the first non-linear x-ray phenomenon that will be explored in good qualitative and quantitative detail, i.e. shows the predictive power of theory.

Summary

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Two main avenues towards real quantum dynamics:

… to describe (time-dependent) ionization dynamics of strongly coupled systems

- "Wave-packet dynamics": Solution of time-dependent Schrödinger equations

  (atomic/molecular physics approach)

- Coherent state evolution: Generalized Bloch-type equations

  (quantum optical approach)

Summary … and where to go ?

- Intense FEL pulses lead to „hollow atoms“ and ions, i.e. extreme matter in a truly exotic form. Dynamics of strongly correlated electrons in controlled light fields is a presently emerging field and require the coherent time evolution of the systems.
- For atoms & molecules, multiphoton ionization and autoionization is likely the first non-linear x-ray phenomenon that will be explored in good qualitative and quantitative detail, i.e. shows the predictive power of theory.

→ Ionization and recombination are indeed fundamental for discovering new phenomena and for obtaining a quantitative understanding of light-matter interactions, both in strong-fields physics and many-particle dynamics.