Dichroism and resonances in intense radiation fields

Tommaso Mazza
SQS Scientific Instrument
European X-Ray Free Electron Laser Facility GmbH

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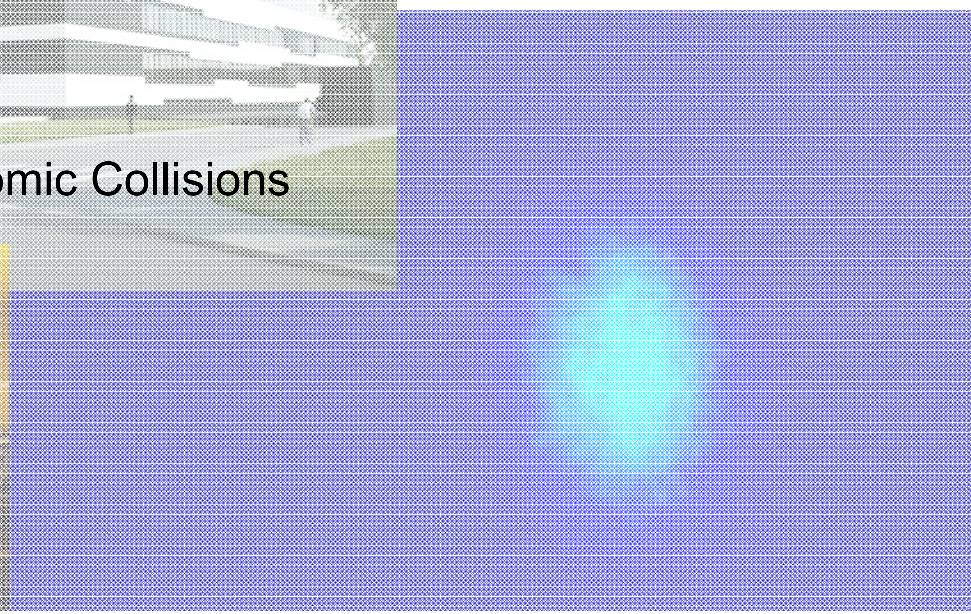
International Conference on Photonic Electronic and Atomic Collisions
ICPEAC30



Pics:
www.xfel.eu
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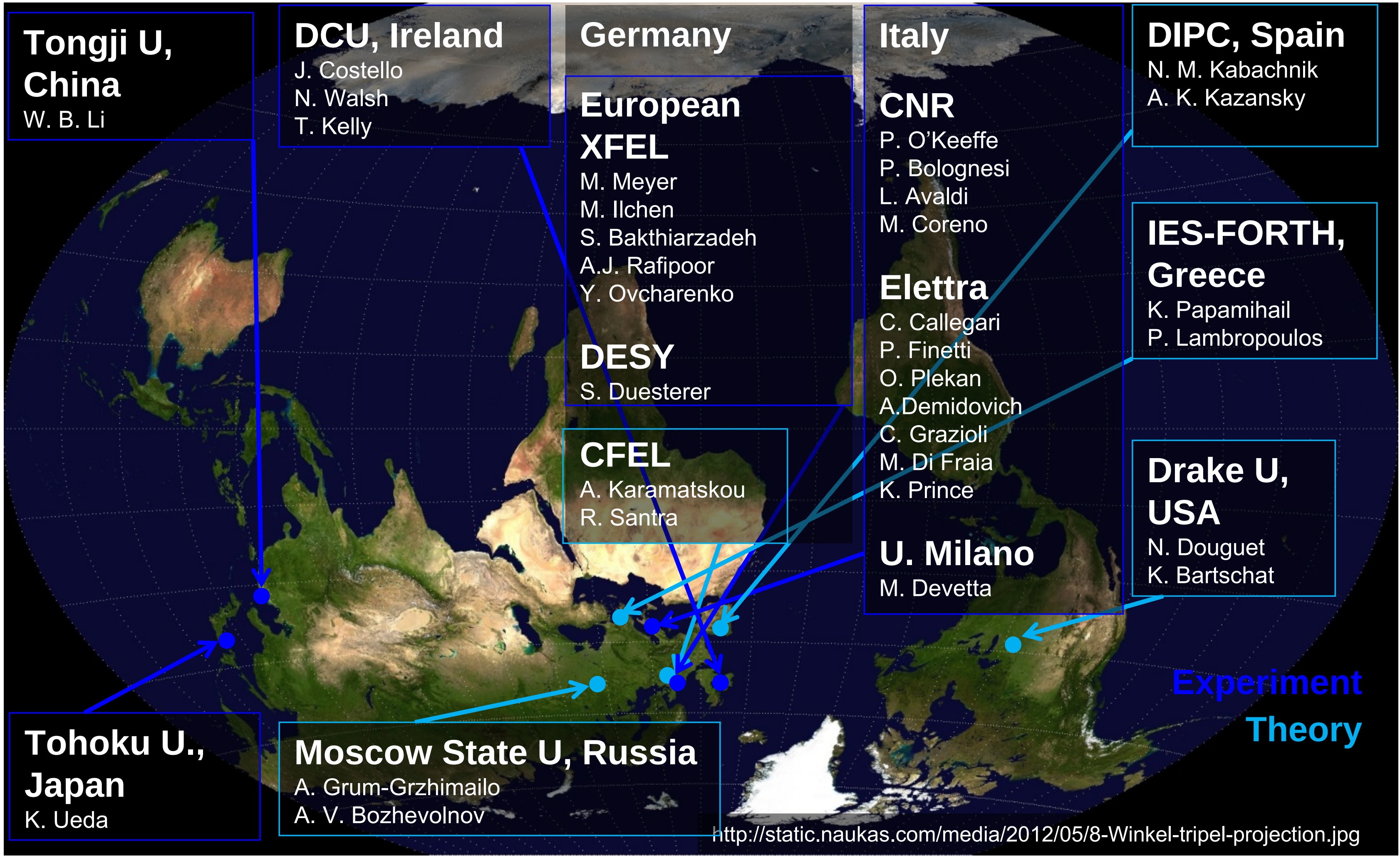
People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People

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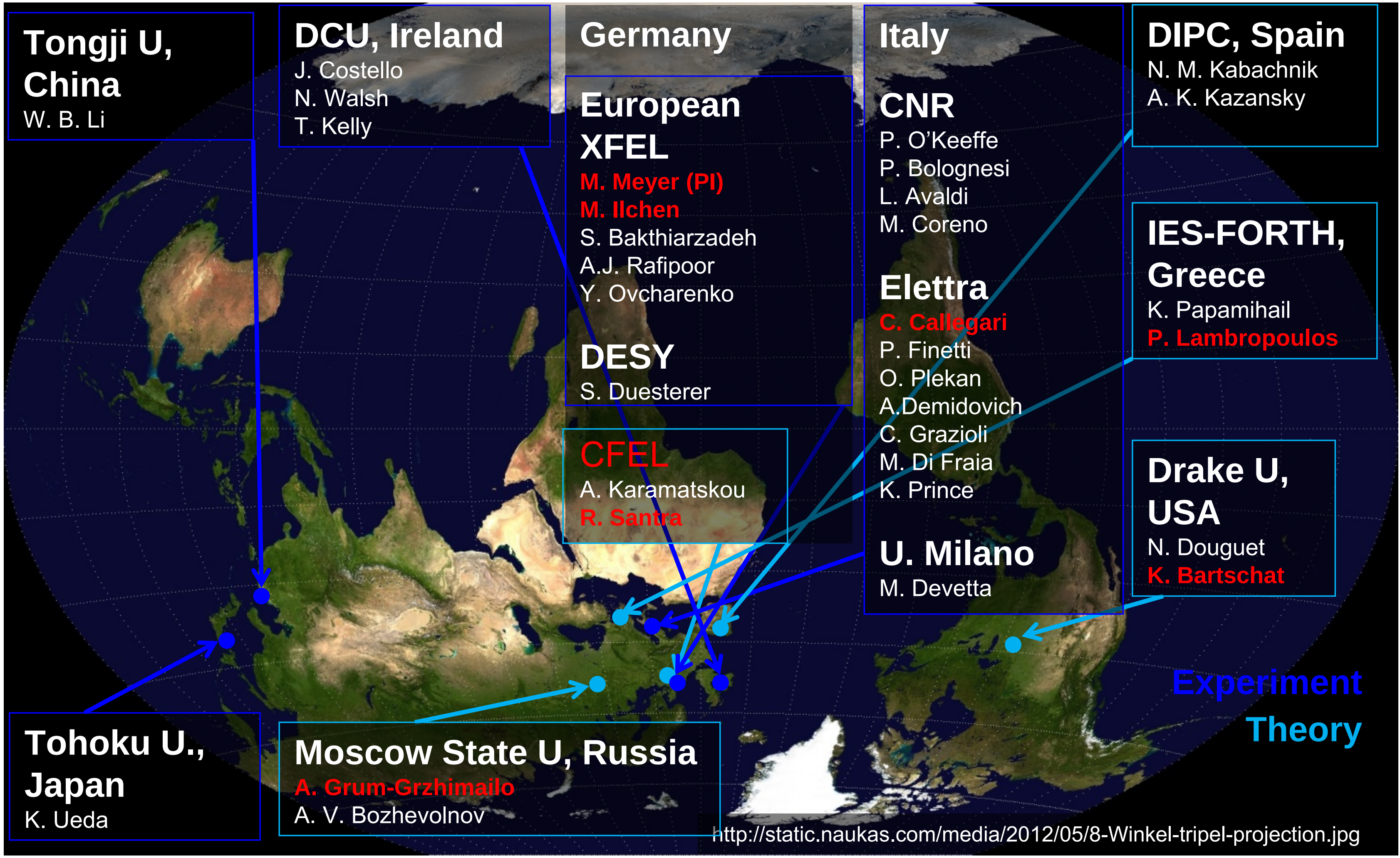
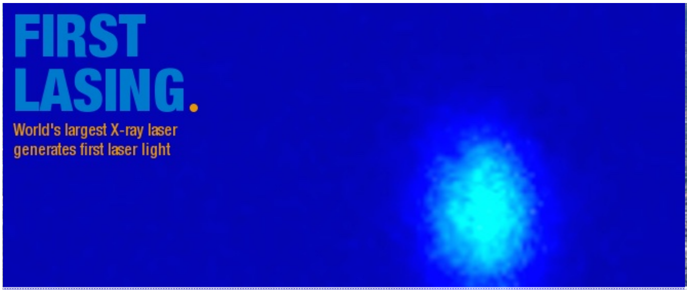
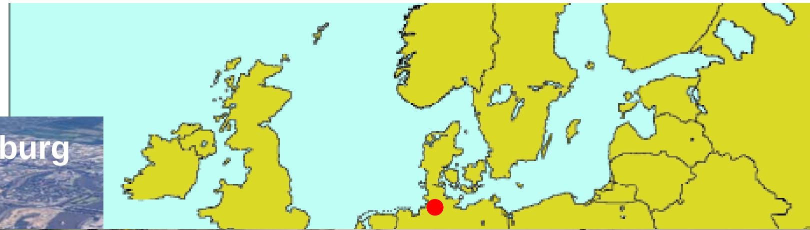
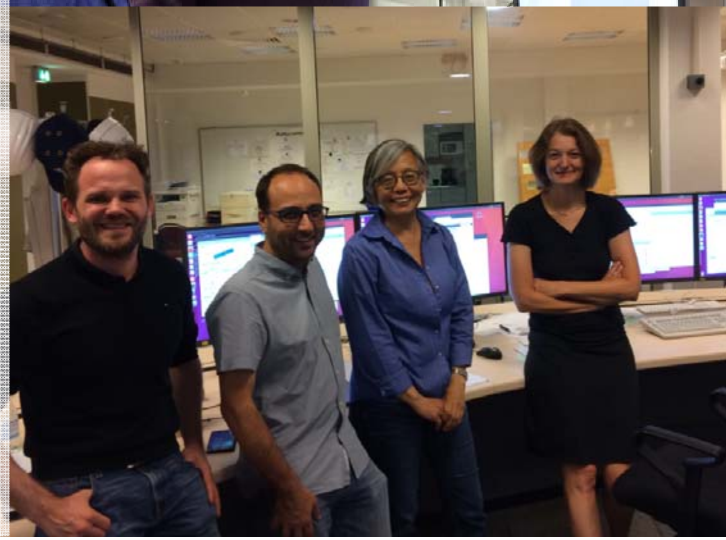
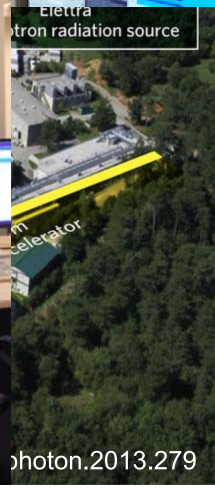


FELs in Europe: present and (very near) future





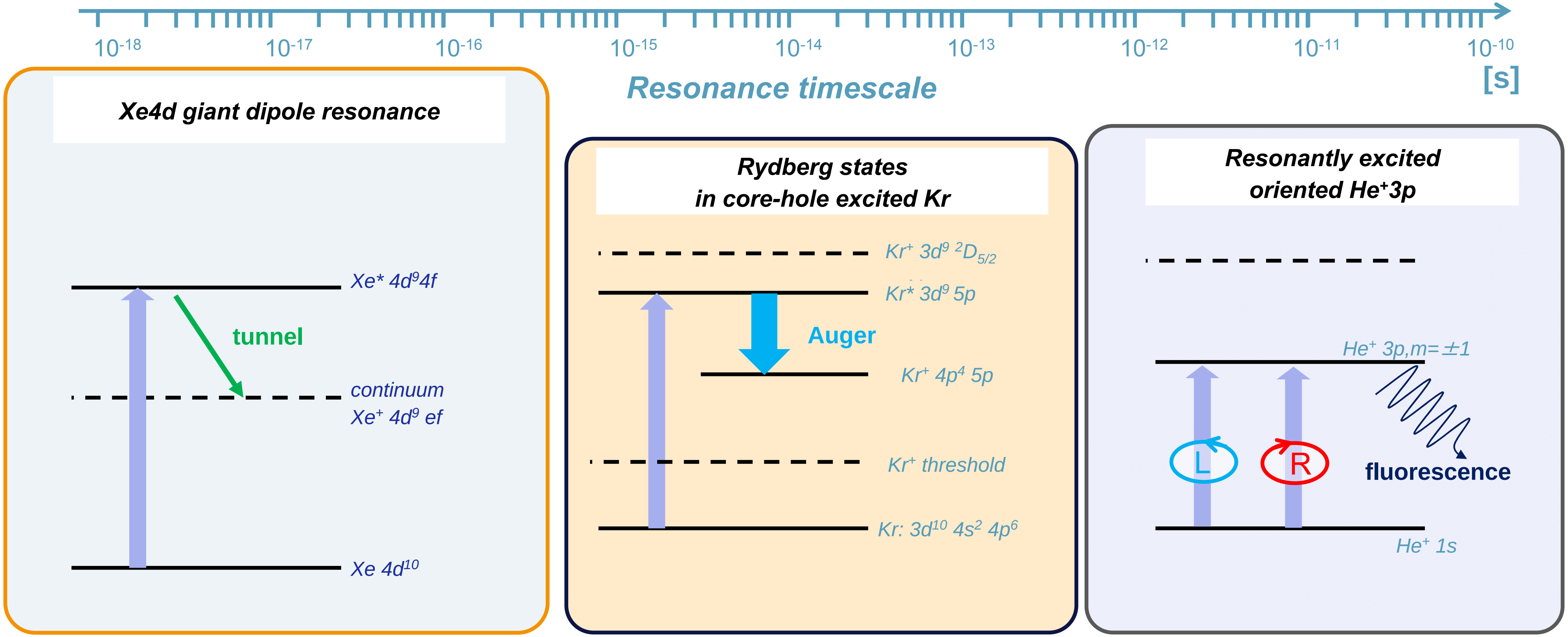




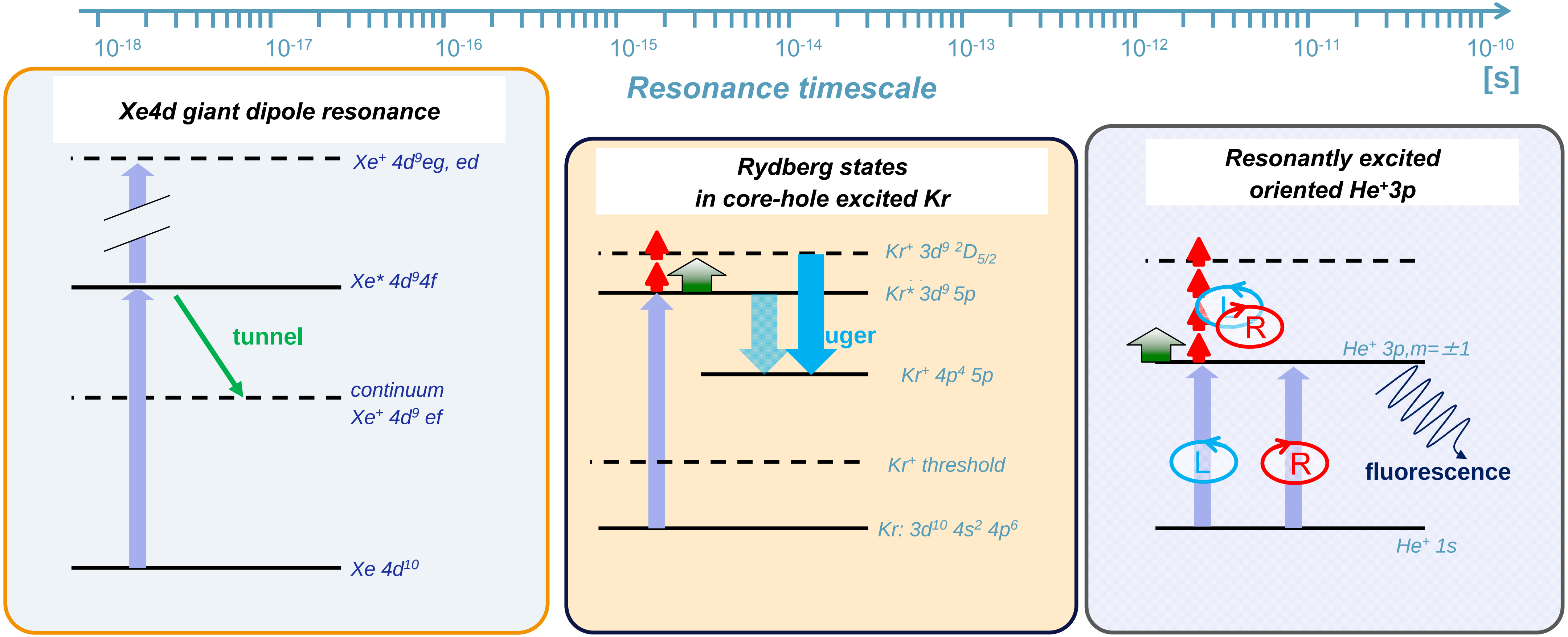
Few facts:

- Lasing at 0.2nm (6.2keV), 1mJ achieved on June 19th;
- Divergence and pointing stability within design specs;
- **Lasing under saturation conditions achieved on July 25th;**
- Commissioning of the instruments on SASE1 (SPB/SFX and FXE) is ongoing right now; first users come in September 2017;
- SQS Scientific Instrument users workshop in November 2017

Dichroism and resonances in intense radiation fields



Dichroism and resonances in intense radiation fields



Outline

- Introduction
- Probing the spectral structure of a collective resonance by nonlinear XUV spectroscopy
- core hole relaxation dynamics control via optical fields
- control of resonant excitation dichroism by optical fields

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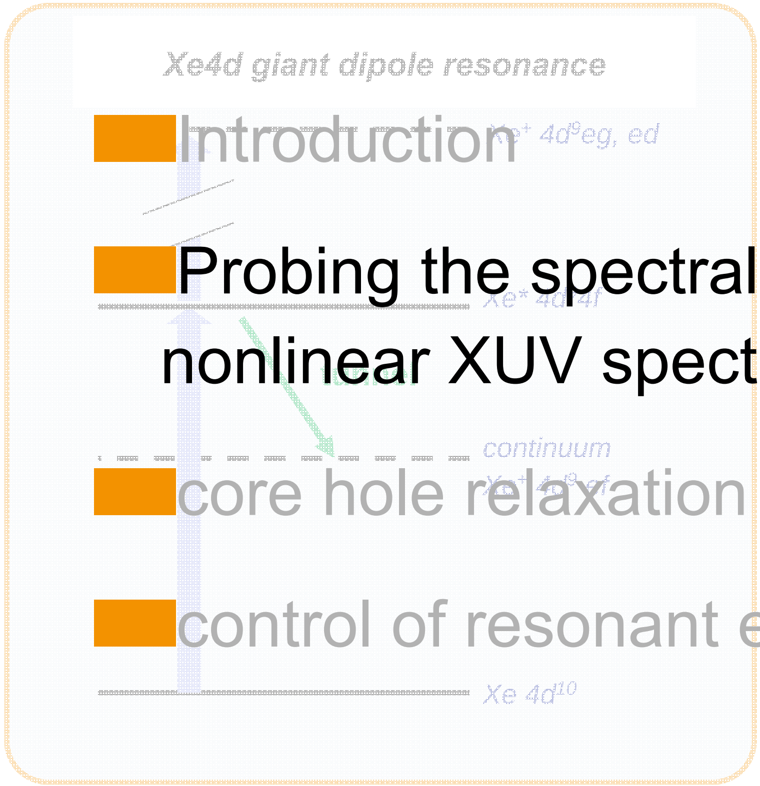
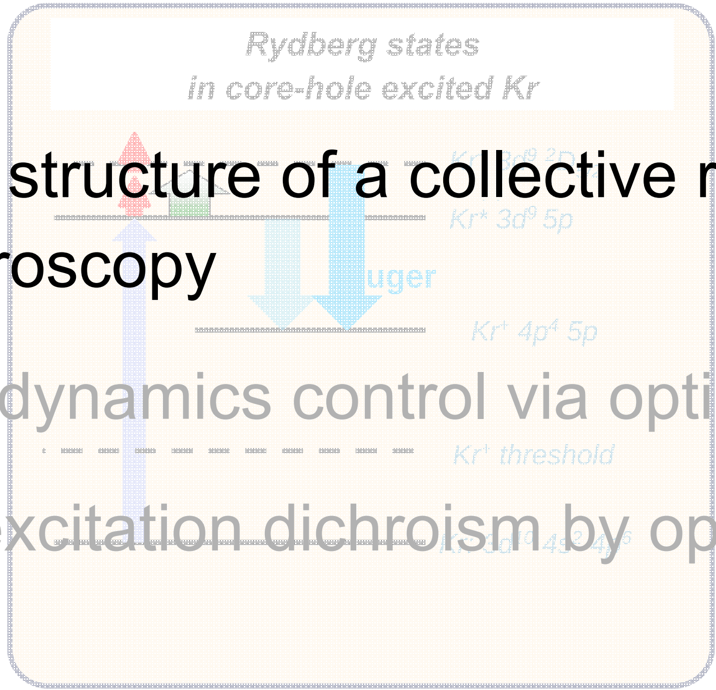
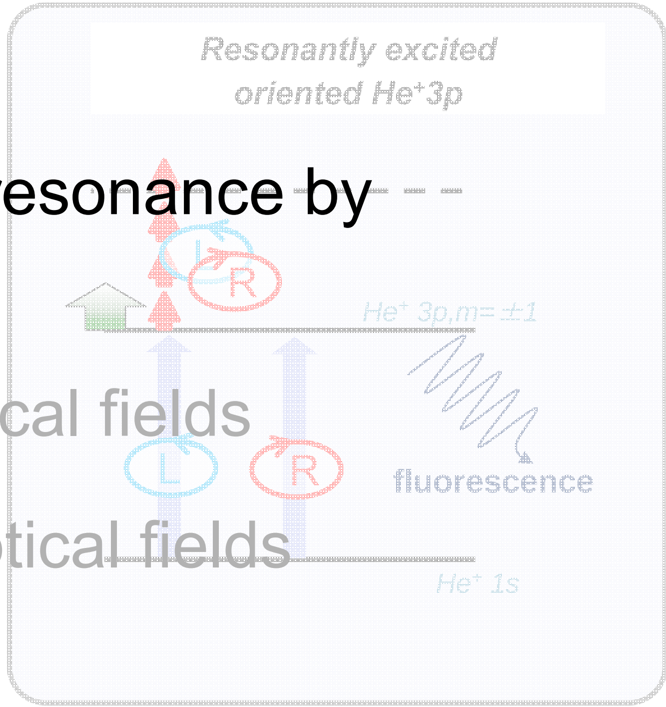
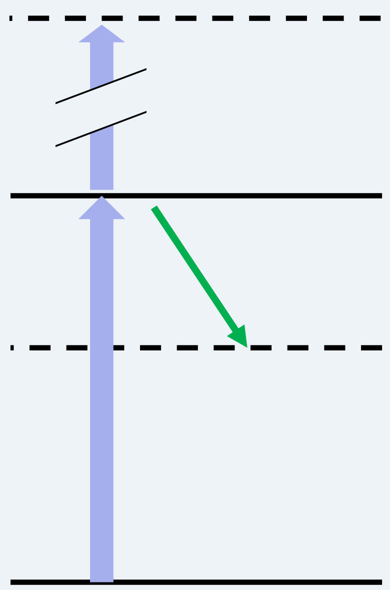






Resonances in intense photon fields: (one color, linearly polarized light) case 1



"How the spectral structure of a collective resonance can be probed by non-linear XUV spectroscopy"

T. Mazza et al., Nature Comms. 6, 6799 (2015)

Xe giant dipole resonance (GDR) *ab initio* theory seen (understood) by an experimentalist

R. Santra, A. Karamatskou, Y.-J. Chen, S. Pabst
PRA 91, 032503 (2015)
J. Phys. B 50, 013002 (2017)

- Shape resonance effect, due to the centrifugal barrier (*l* = 3) the electron promoted to the continuum is trapped in a resonant state before tunneling out;
- Only when **electron correlation effects** within the 4d shell **are included** we get quantitative agreement with experimental data
- How to include electron correlation effects by TDCIS:

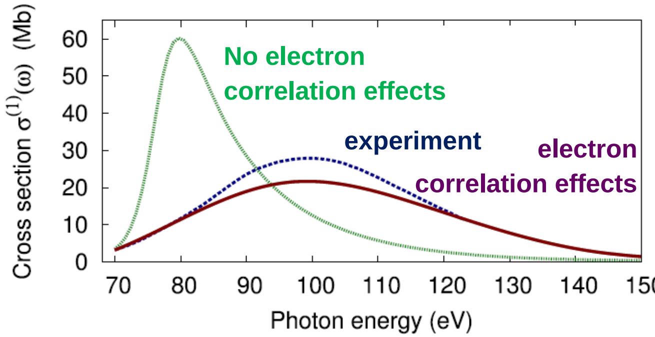


Figure 11. One-photon absorption cross section of xenon calculated within TDCIS using the two models. The experimental curve [68] resembles the interchannel curve [69].

A. Karamatskou, J. Phys. B: 50 (2017) 013002

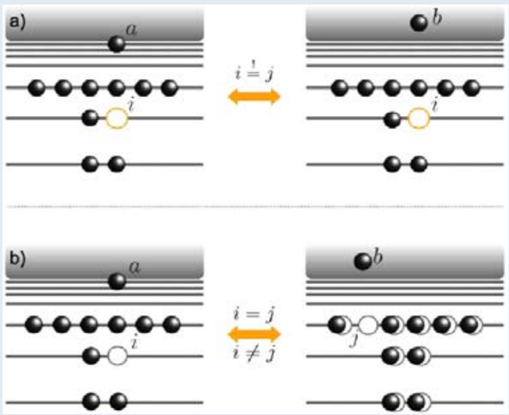
$$\hat{H} = \sum_{n=1}^{N}\left(\frac{\hat{\mathbf{p}}_n^2}{2} - \frac{Z}{|\hat{\mathbf{r}}_n|} \right. \ldots$$

$$+ \left(\frac{1}{2}\sum_{\substack{n,n'=1 \\ n \neq n'}}^{N} \frac{1}{|\hat{\mathbf{r}} \ldots} \right. \ldots$$

$$=: \hat{H}_0 + \hat{H}_1,$$

"intrachannel coupling" (reduced): the outgoing electron couples only to the hole it originates from

"interchannel coupling" (full): the outgoing electron is coupled to all possible hole states



Xe giant dipole resonance (GDR) ATI *ab initio* theory seen (understood) by an experimentalist

R. Santra, A. Karamatskou, Y.-J. Chen, S. Pabst
PRA 91, 032503 (2015)
J. Phys. B 50, 013002 (2017)

Consequences on the predicted physics of the interchannel coupling inclusion (full model):

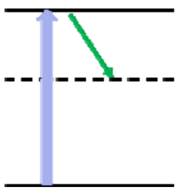
1.The Xe4d GDR is described as a superposition of particle-hole states, i.e. it is a truly collective effect;

2.Non-degenerate poles in the resonance structure are predicted

	SES[a]	
	Ξ_n (eV)	Γ_n (eV)
Intrachannel		
$4d_0$	76.3	8.3
$4d_{\pm1}$	77.6	13.8
$4d_{\pm2}$	77.2	10.6
Full CIS		
R_1	74.3	24.6
R_2	107.2	59.9

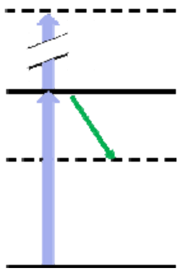
[a]All SES values have an error bar of 0.1 eV. This is ca

Chen et al., PRA **91**, 032503 (2015)



$$\sigma^{(1)} = \left|\sum_F \frac{\langle F|\hat{H}_{int}|I\rangle}{E - E_F + \frac{i}{2}\Gamma_F + }\right.$$

1-photon cross section:
resonance final states are not resolved



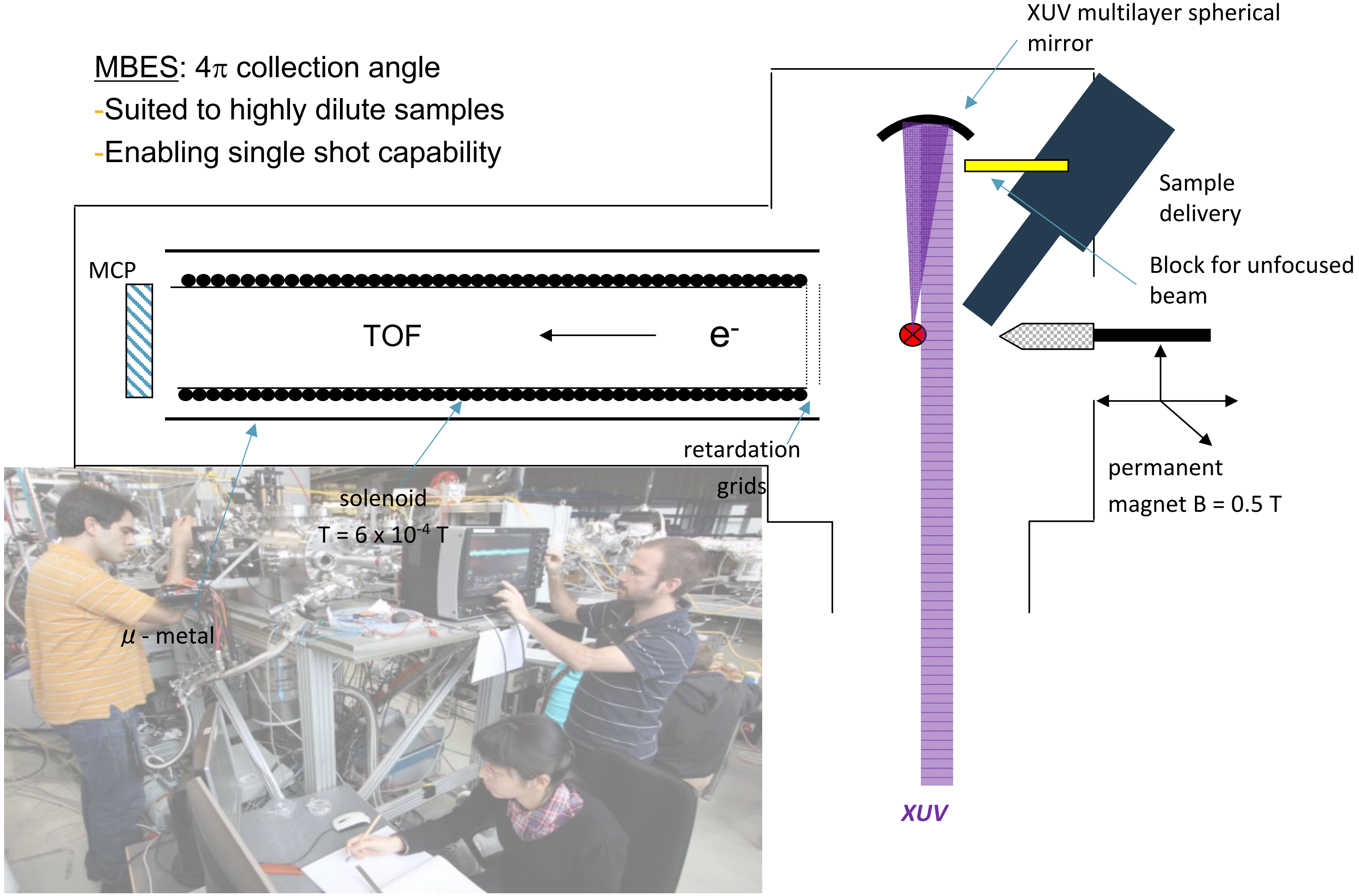
$$\sigma^{(2)} = \left|\sum_{M_{res}} \frac{\langle F|\hat{H}_{int}|M_{res}\rangle\langle M}{E - E_{M_{res}} + \frac{i}{2}}\right.$$

***2-photon ATI cross section**:*
*Interference between overlapping resonances arise, whose relative phase **can** change the shape of the cross section curve*

XUV non-linear Spectroscopy at FLASH: Experimental apparatus

MBES: 4π collection angle

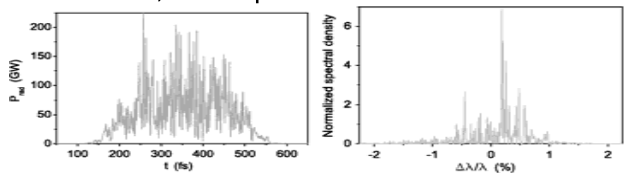
- Suited to highly dilute samples
- Enabling single shot capability



XUV source: FLASH

hv = 105, 140 eV

BW ~< 1eV, SASE process



Schneidmiller et al., J. Micro/Nanolith. MEMS MOEMS. 11(2), 021122

Pulse duration ~ 50-100 fs,
focus ~3-5 um, gaussian model

→ 10 uJ ~ 10^{14} - 10^{15} W/cm^2

MBES:

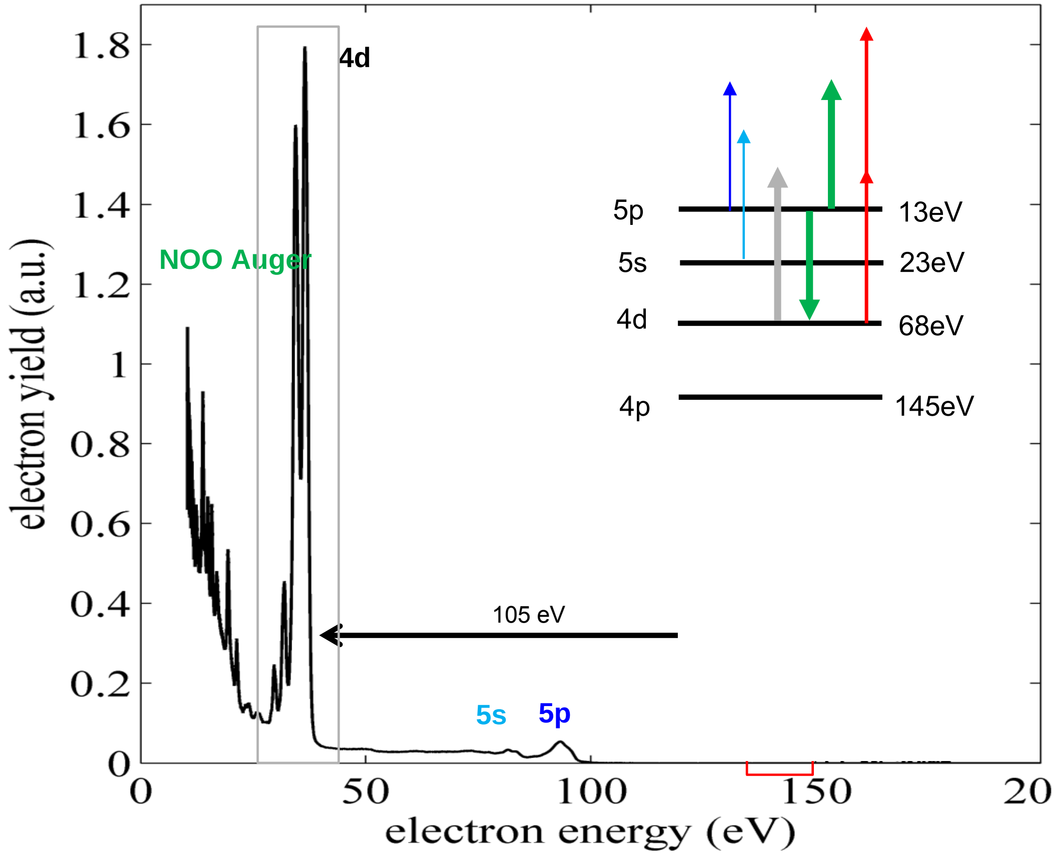
ΔKE / KE ~ 2% on ret. electrons

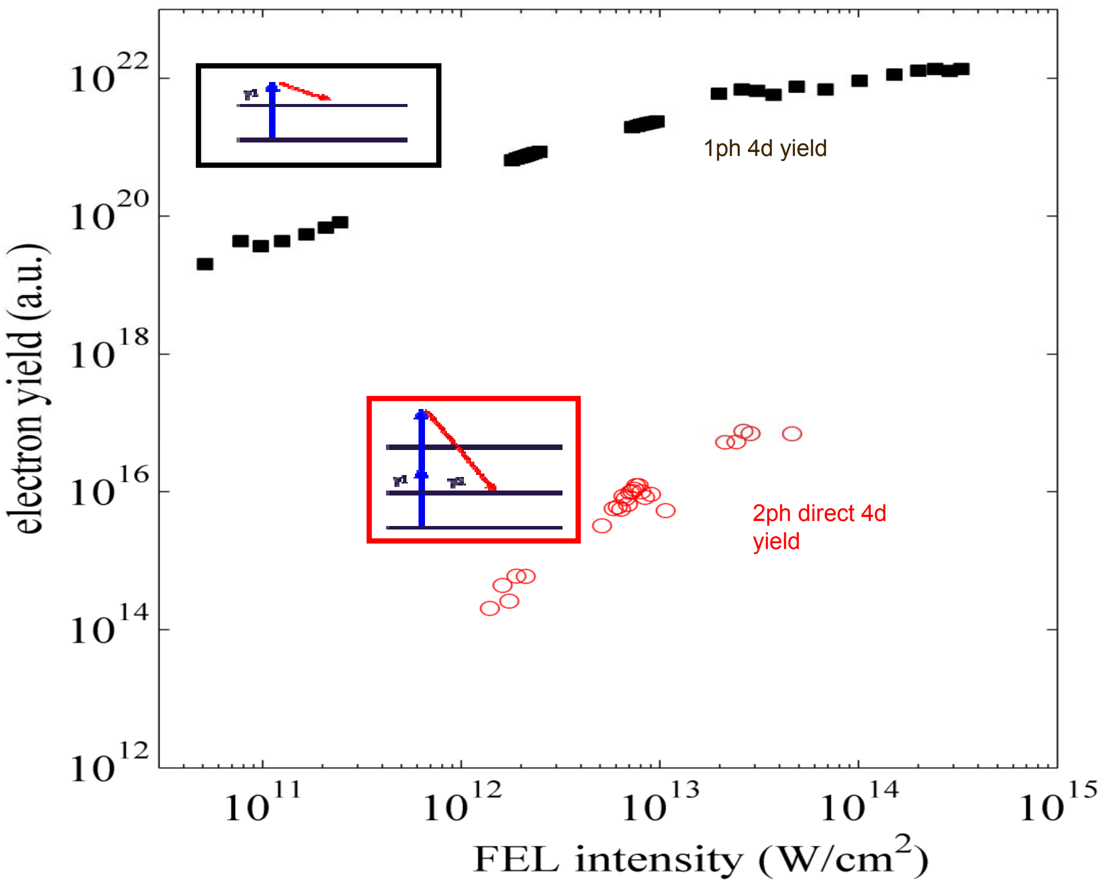
Single shot capability (4π acceptance)

Sample delivery:

Sample density can be tuned over ~2 o.m. in a controlled way

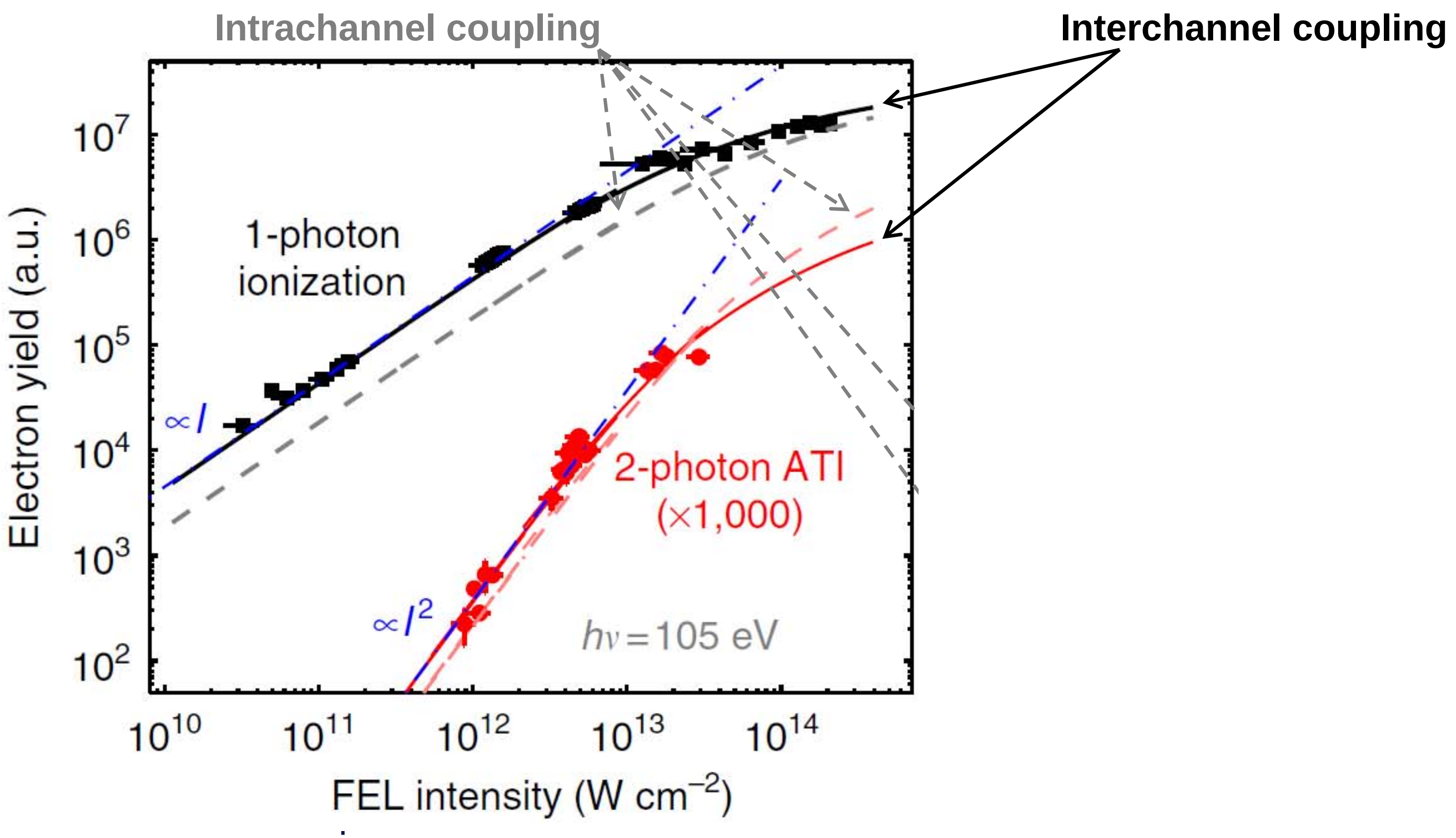
Above Threshold Ionization of Xe4d at hv = 105eV





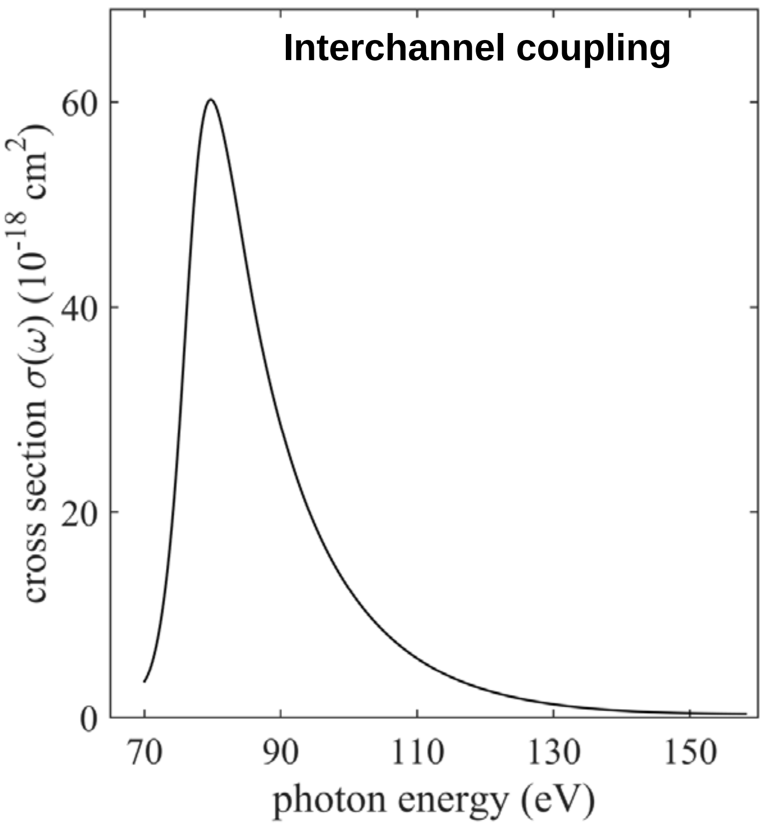
T. Mazza et al., Nature Comms. 6, 6799 (2015)

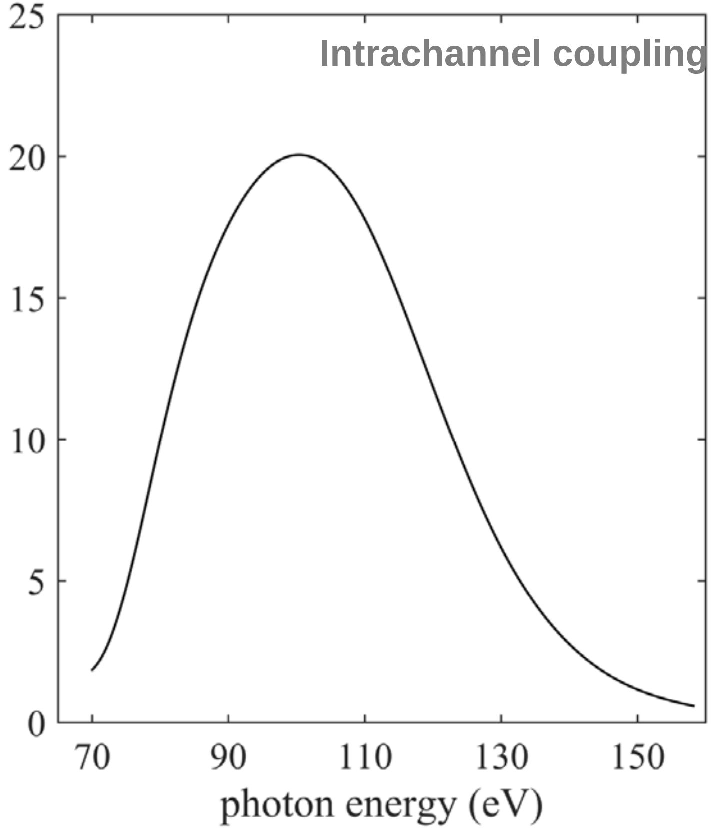
Above Threshold Ionization of Xe4d at hν = 105eV, 140eV: power law comparison between experiment and theory



T. Mazza et al., Nature Comms. 6, 6799 (2015)

TDCIS of the 1-photon and 2-photon Xe4d ionization





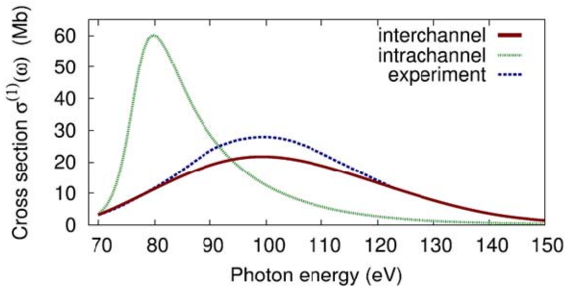


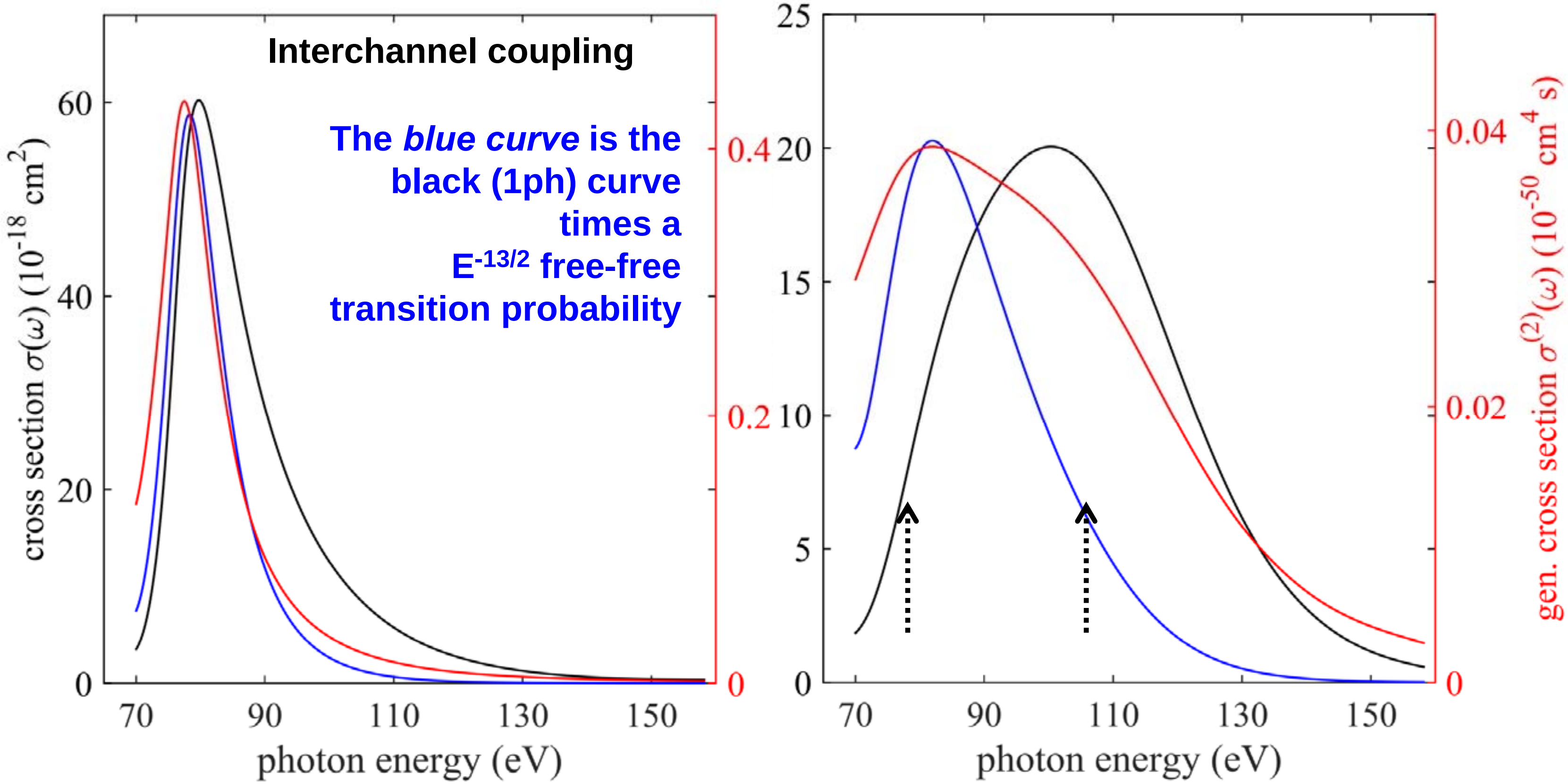
Figure 11. One-photon absorption cross section of xenon calculated within TDCIS using the two models. The experimental curve [68] resembles the interchannel curve [69].

A. Karamatskou, J. Phys. B: 50 (2017) 013002

Interchannel vs. intra σ(ω)
- blue-shift
- broadening

T. Mazza et al., Nature Comms. 6, 6799 (2015)

TDCIS of the 1-photon and 2-photon Xe4d ionization



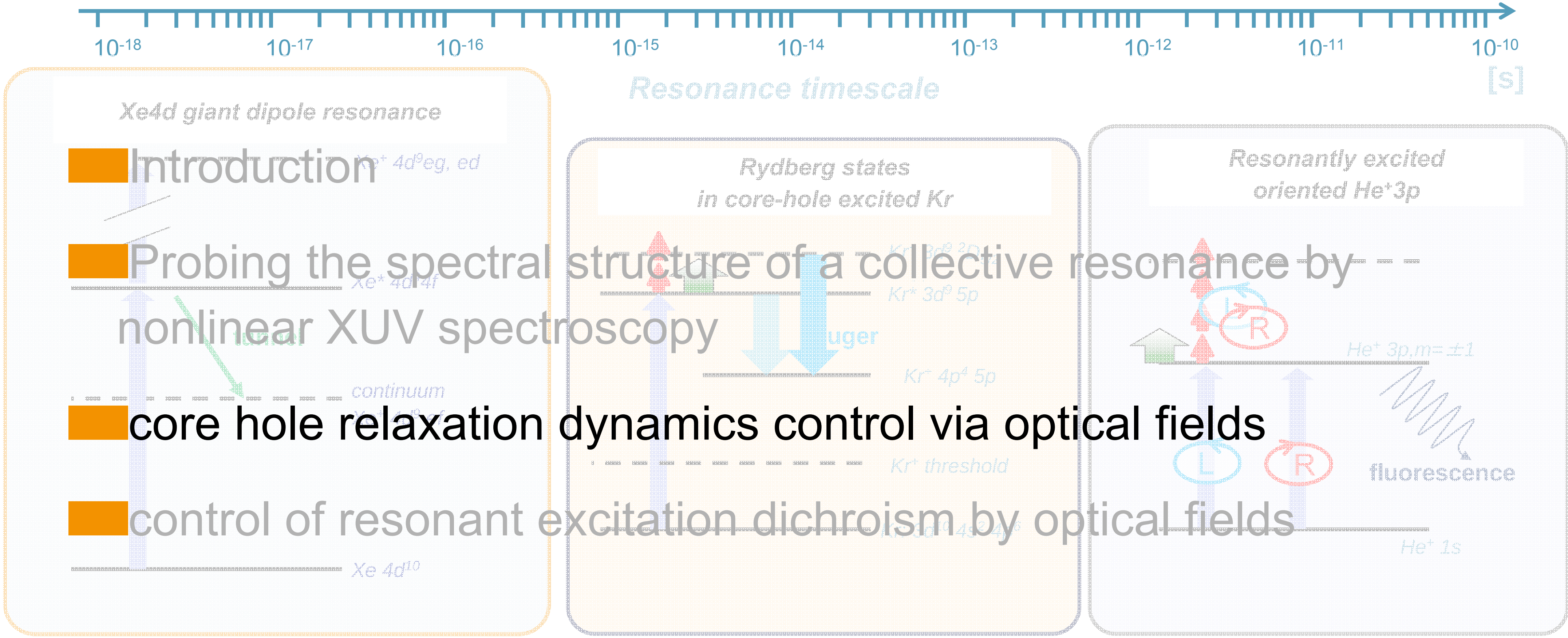
Interchannel vs. intra σ(ω)
- blue-shift
- broadening

Intra σ(ω) vs. $\sigma^{(2)}(\omega)$
- ➡ red-shift and sharpening
- ➡ Consistent with 2-factorization

Inter σ(ω) vs. $\sigma^{(2)}(\omega)$
- ➡ red-shift and **broadening**

T. Mazza et al., Nature Comms. 6, 6799 (2015)

Outline

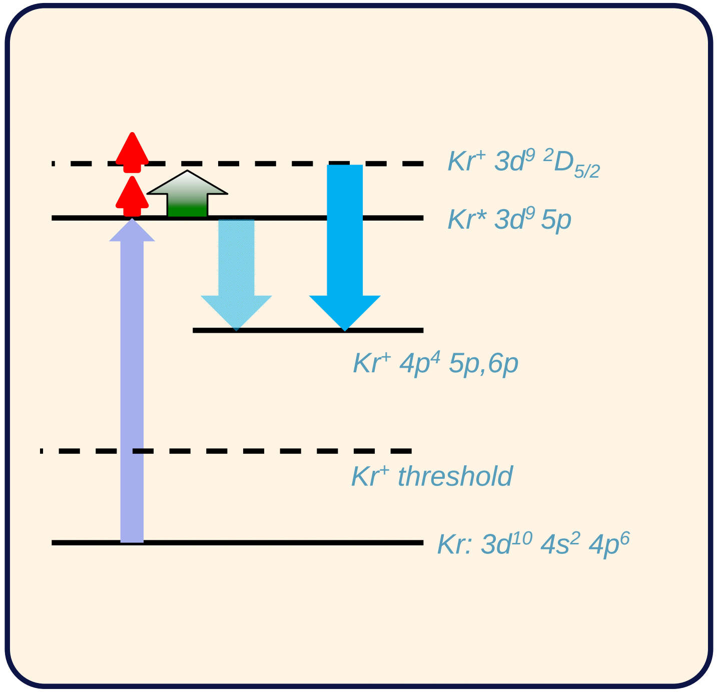


- Introduction
- Probing the spectral structure of a collective resonance by nonlinear XUV spectroscopy
- core hole relaxation dynamics control via optical fields
- control of resonant excitation dichroism by optical fields

Resonances in intense photon fields: (two-color, linearly polarized light) case 2

1. New channel introduced by the MPI ionization of the Kr*5p Rydberg state, competing with the Auger decay
2. AC Stark shift introduced by the IR field

"controlling core hole relaxation dynamics via intense optical fields"



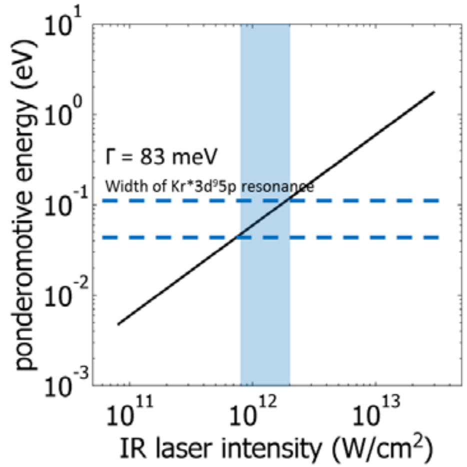
Kr*5p is very close to the threshold

↓

Polarizability ~ $1/\omega^2$

↓

AC Stark shift ~ ponderomotive shift
$Up = eE_0^2 / 4m\omega^2$

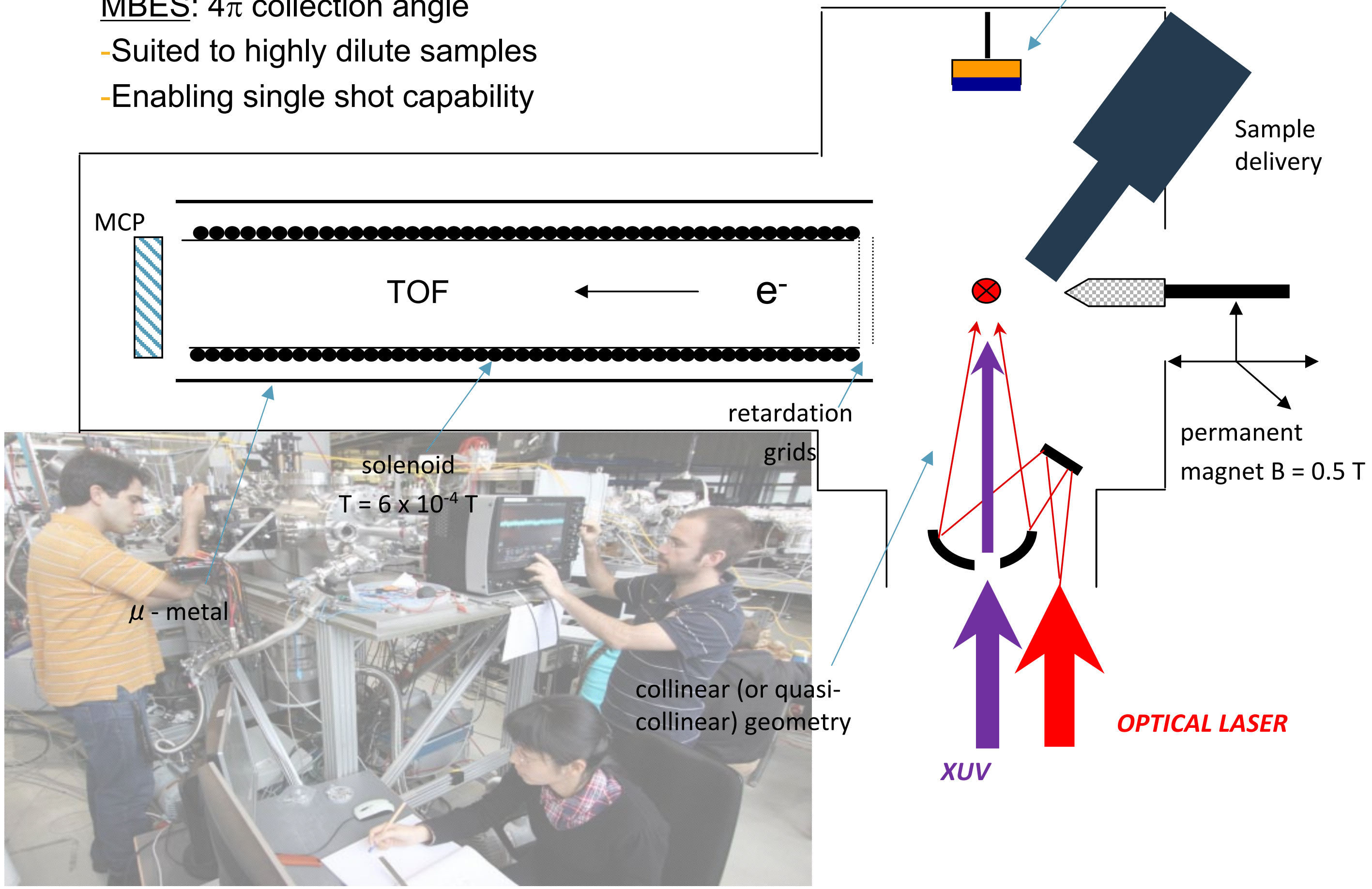


T. Mazza *et al*, *J. Phys. B* **45** 141001 (2012)

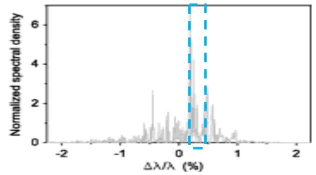
L. B. Madsen, et al., Phys. Rev. Lett. **85**, 42
S. E. Harris, Physics Today **50**, 7, 36 (1997)
Glover, Santra, Young et al., *Nature Physics* **6**, 69 - 74 (2010)

XUV-IR Electron Spectroscopy at FLASH: Experimental apparatus

MBES: 4π collection angle
- Suited to highly dilute samples
- Enabling single shot capability



XUV source: FLASH
hν = 90-92 eV (Kr3d5p resonance)
BW ~< 1eV, **~50meV after mono**



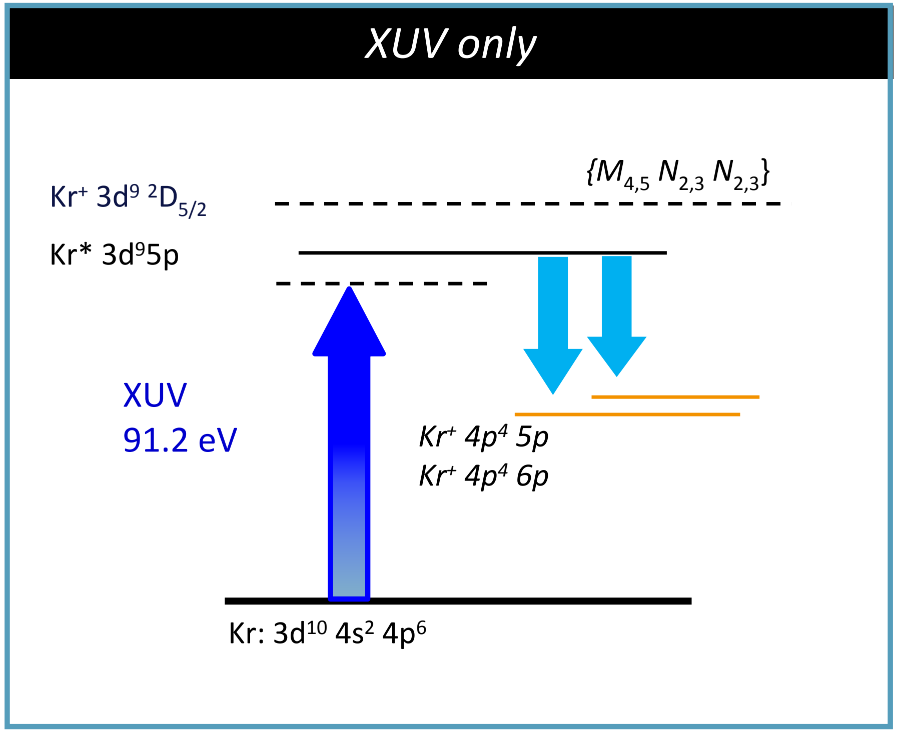
Schneidmiller et al., J. Micro/Nanolith. MEMS MOEMS. 11(2), 021122

Pulse duration ~ 50-100 fs,
Intensity irrelevant

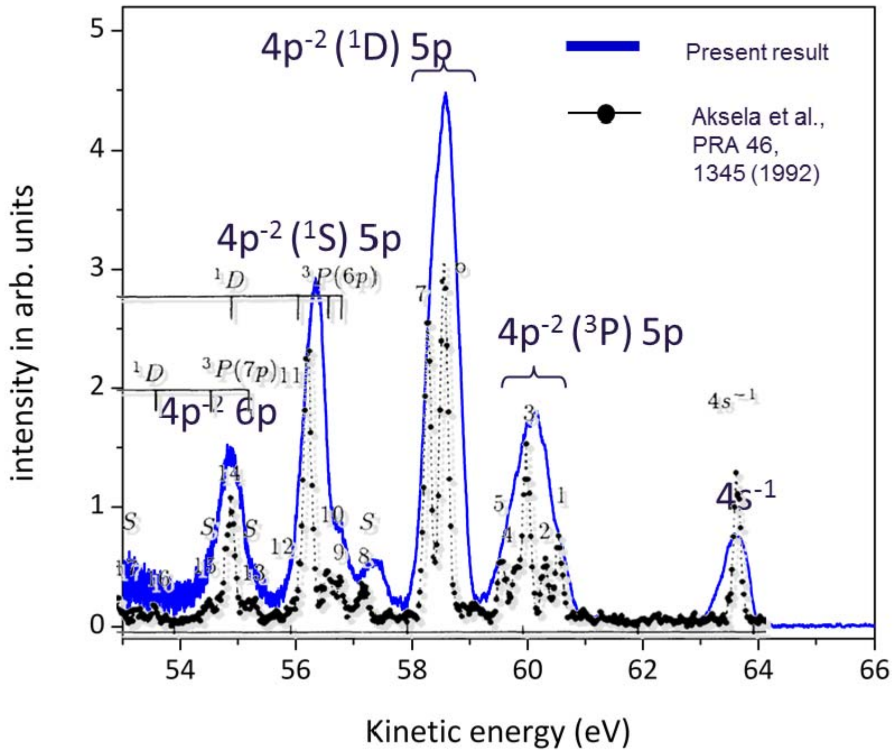
IR:
λ = 800 nm
Intensity ~ 10^{12} W/cm^2
No pump probe: time overlap

MBES:
ΔKE / KE ~ 2% on ret. electrons
Single shot capability (4π acceptance)

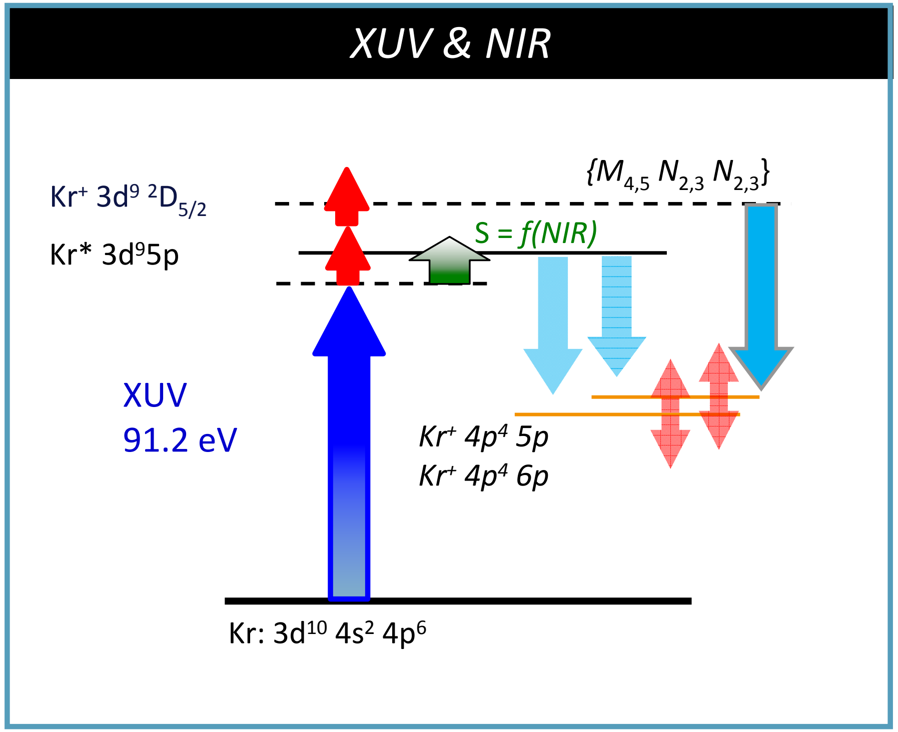
Electron Spectroscopy:
Auger decay of resonantly excited Kr $3d^{-1}$ 5p states

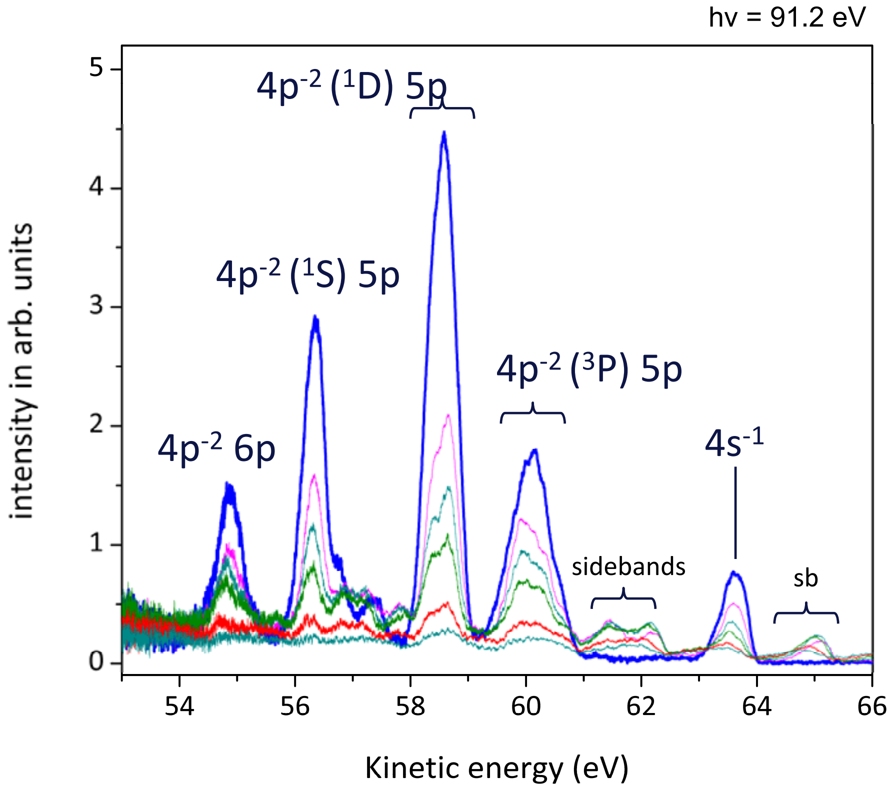


hν = 91.2 eV

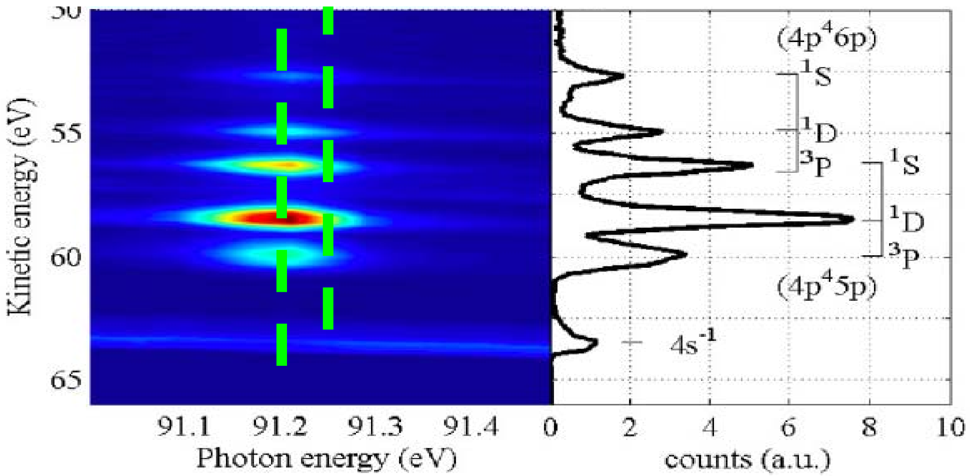


Electron Spectroscopy:
Auger decay suppressed by dressing IR field





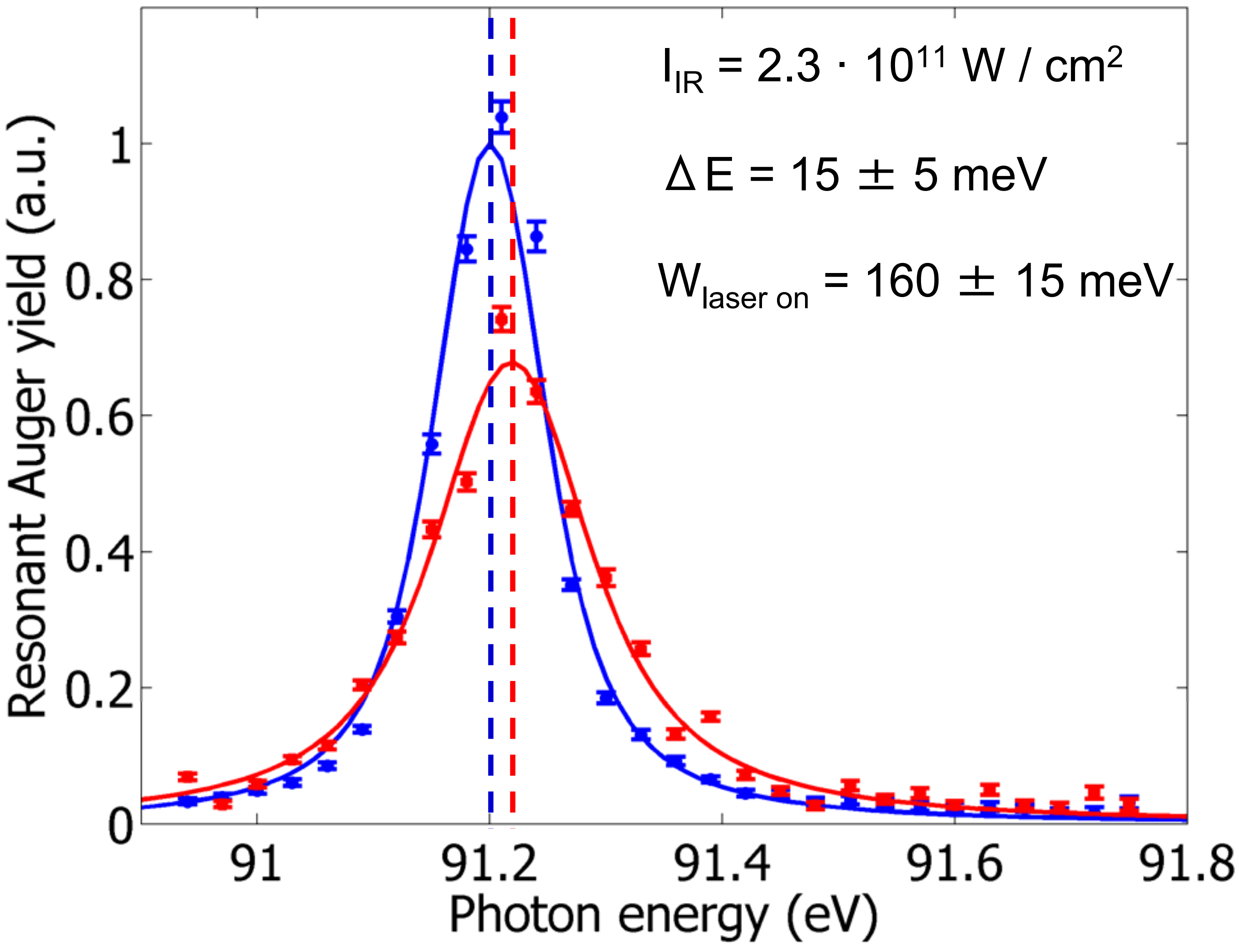
hv-dependent electron yield: influence of the dressing IR field

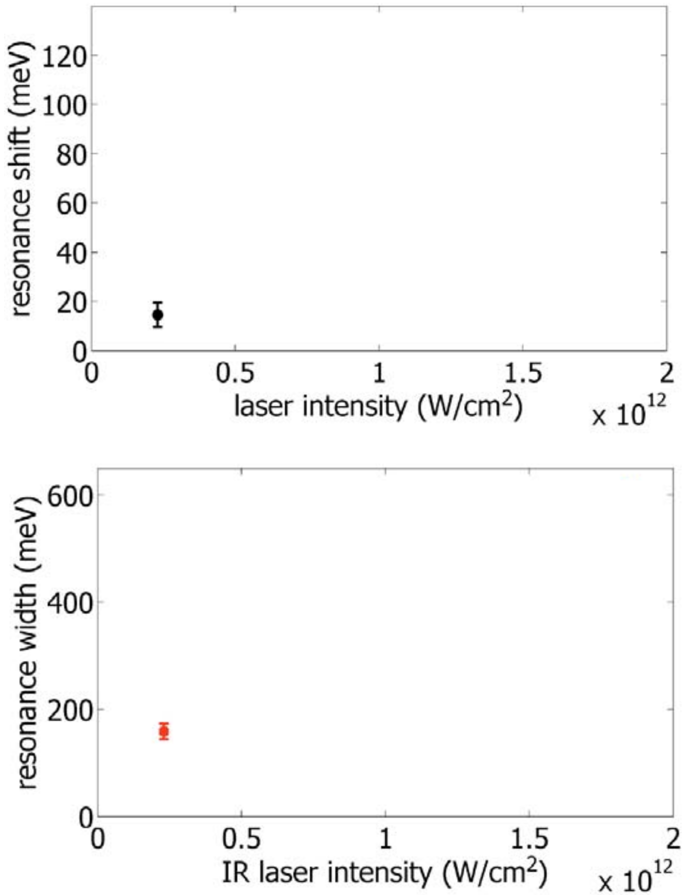


NIR laser OFF

The resonance lineshape is retrieved from the integrated resonant Auger electron yield ***normalized over the 4p PE line yield***

hv-dependent electron yield: influence of the dressing IR field





T. Mazza *et al*, *J. Phys. B* **45** 141001 (2012)

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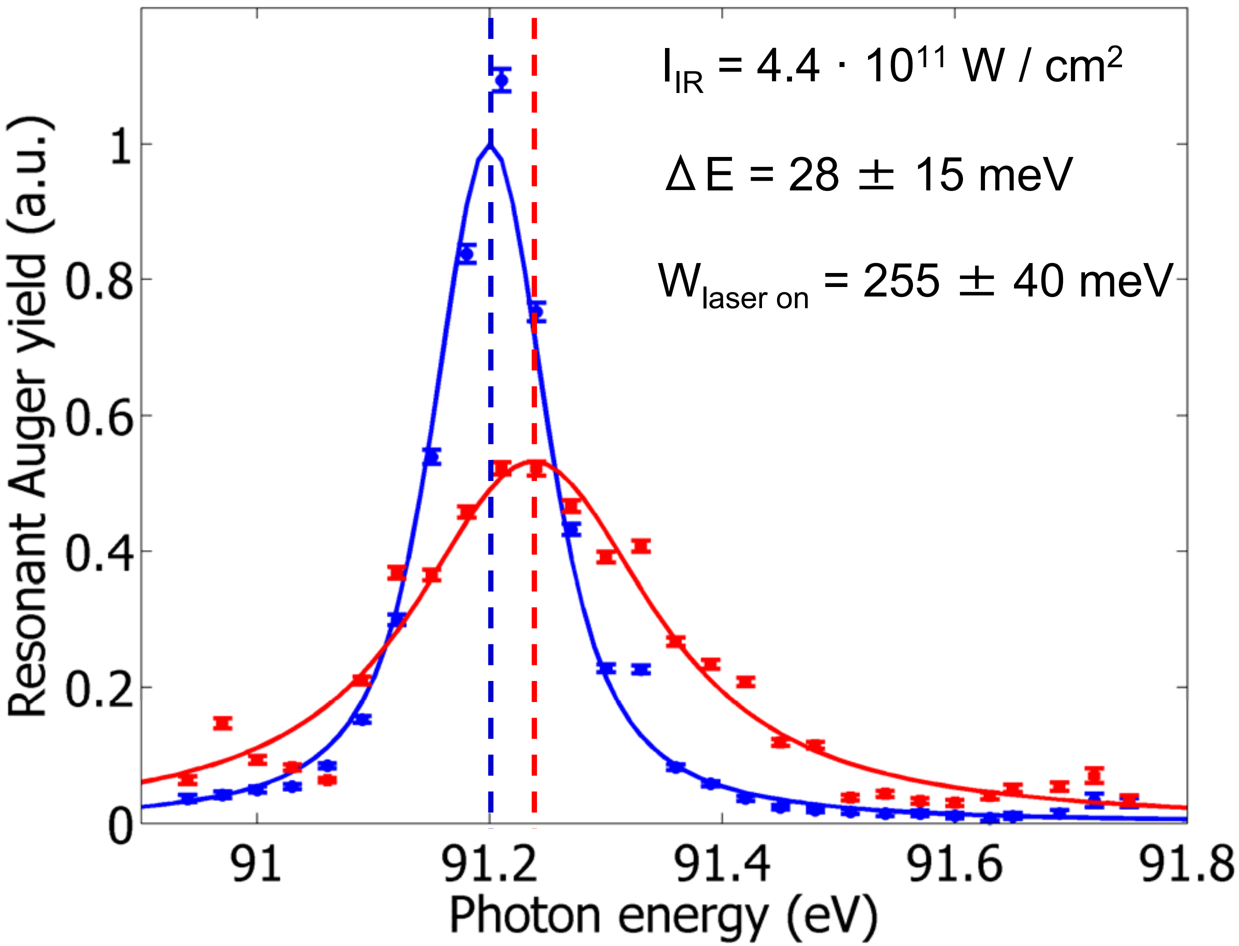

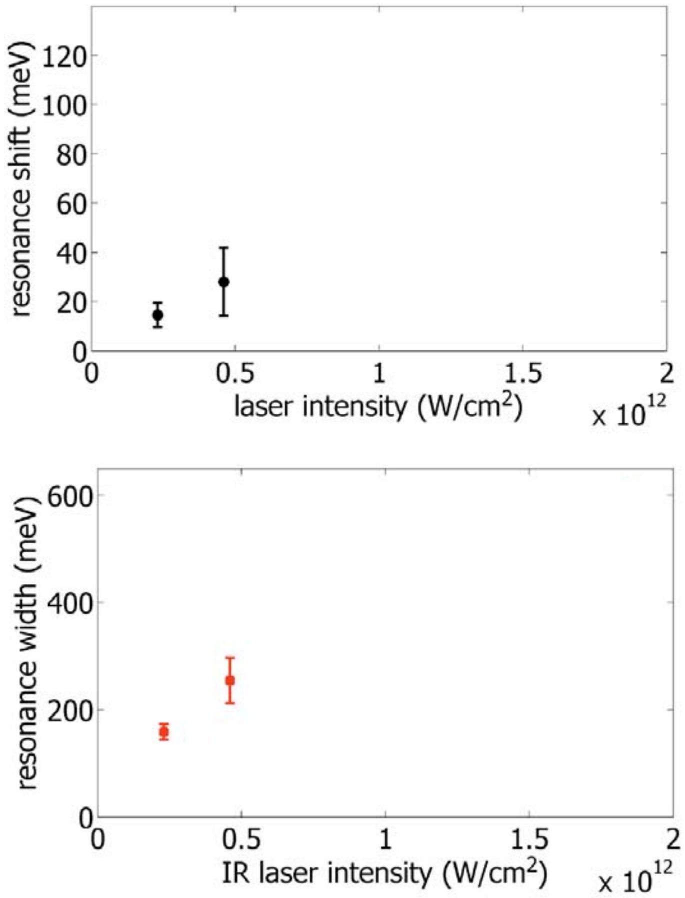

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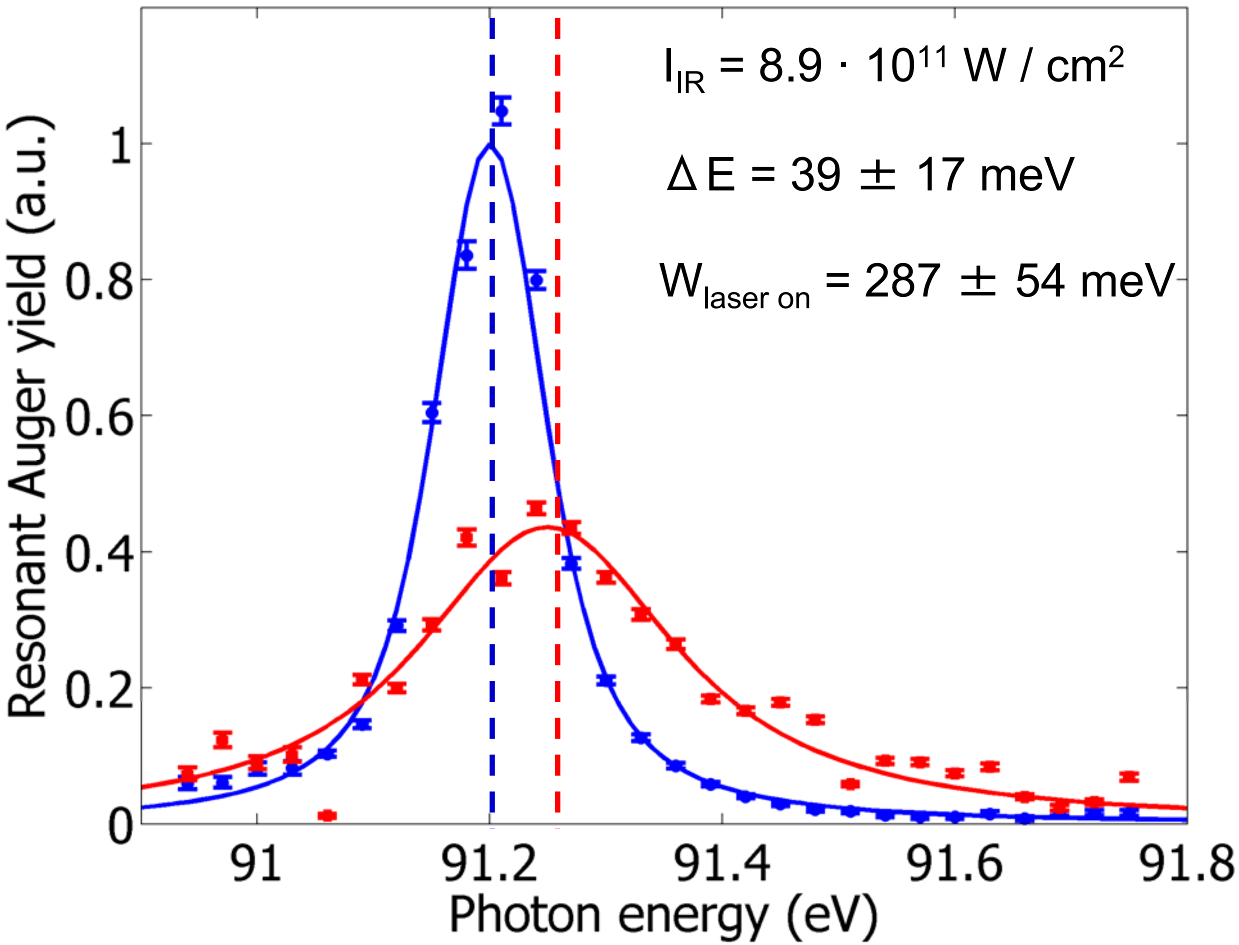

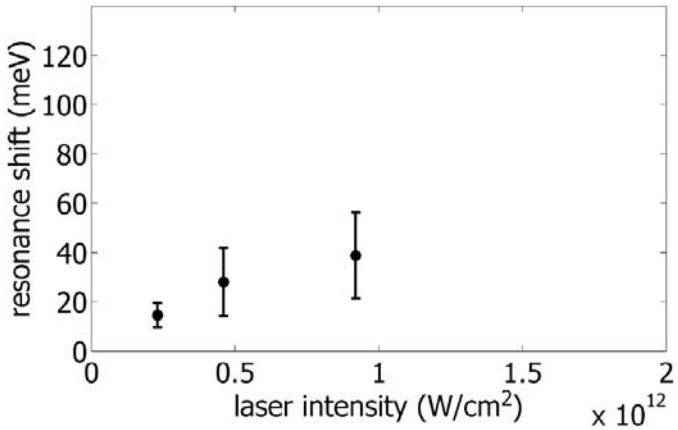

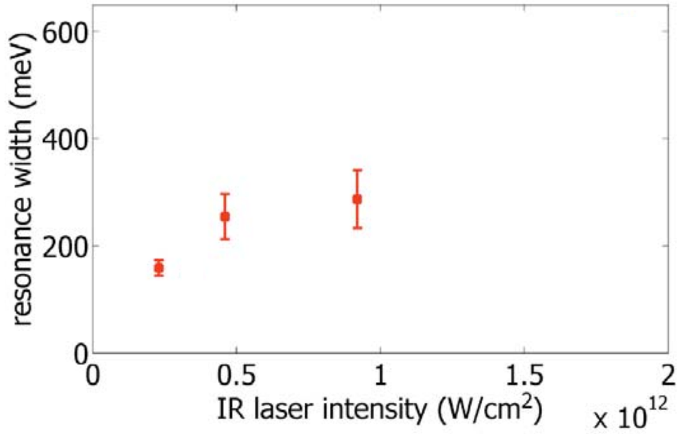

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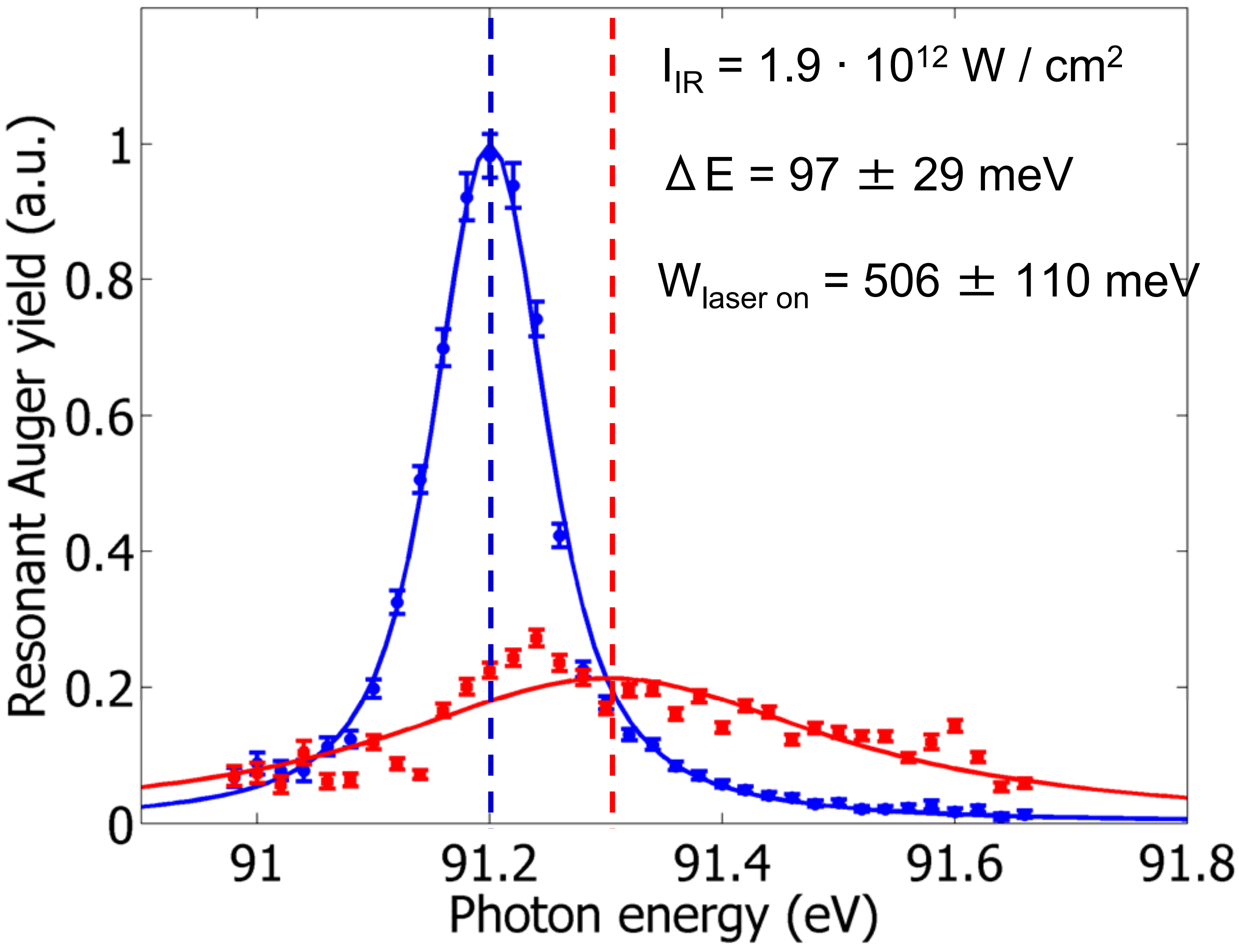
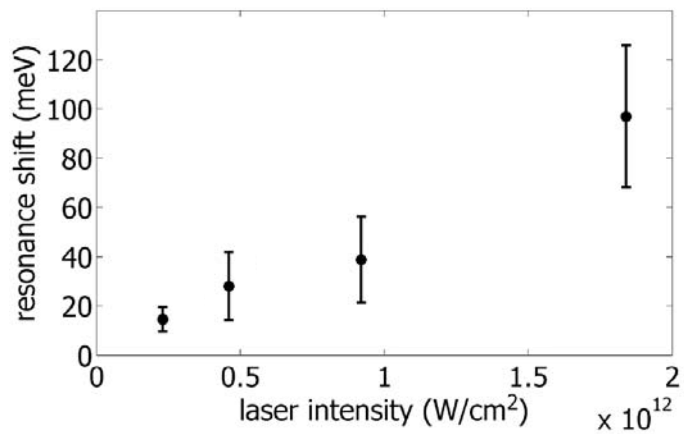
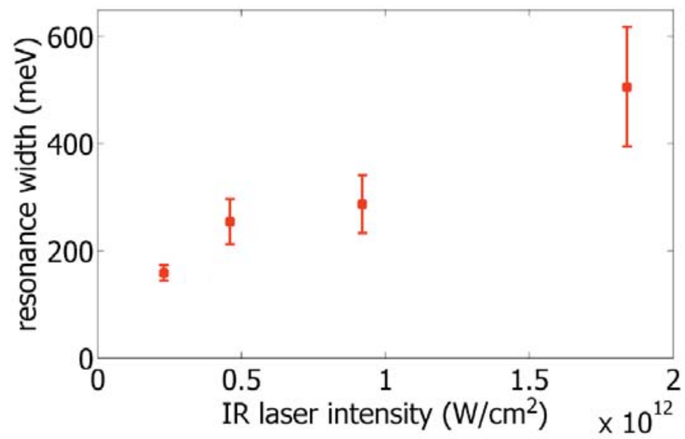




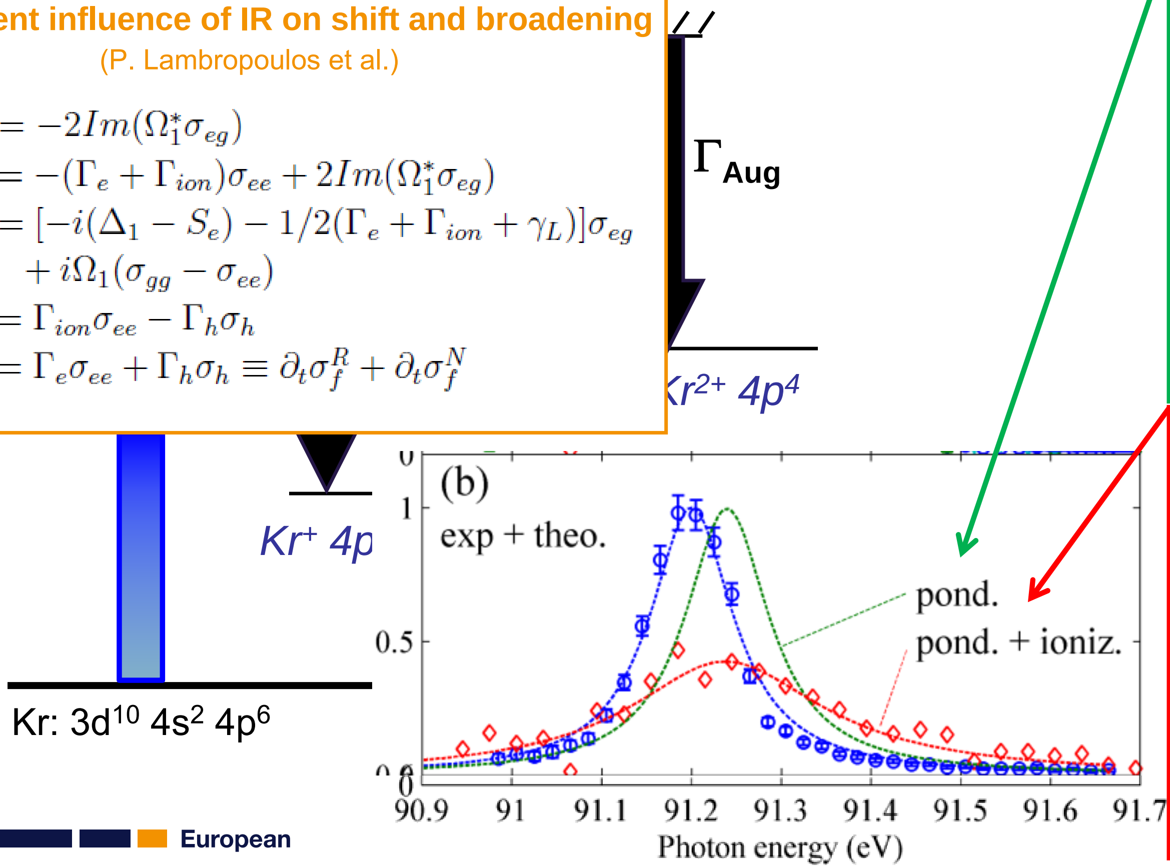


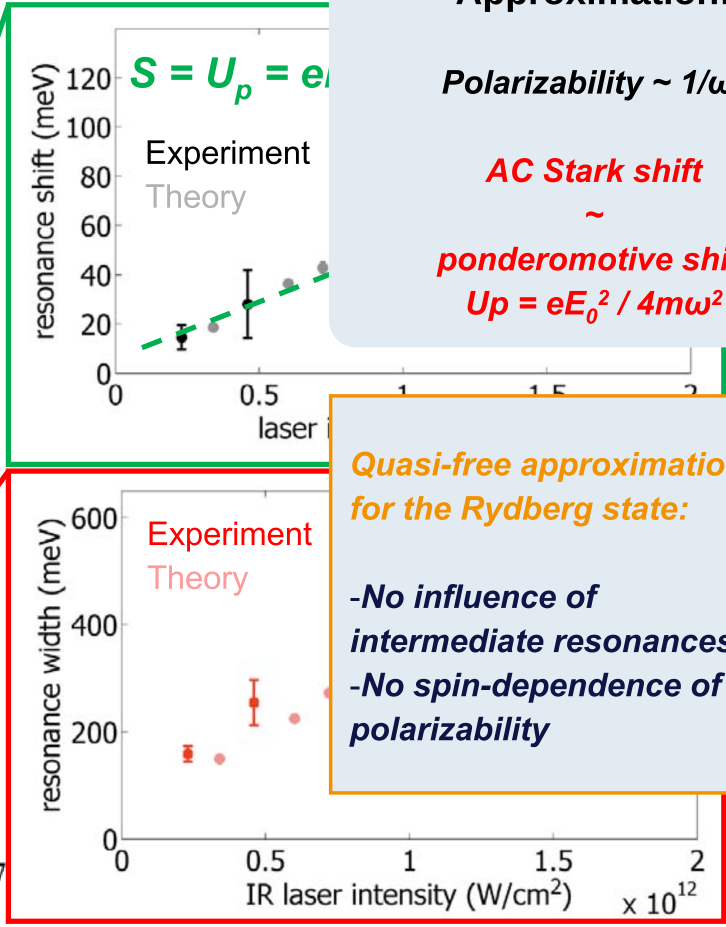
T. Mazza *et al*, *J. Phys. B* **45** 141001 (2012)

Dynamic Stark shift
controlling the relaxation dynamics

Solution of rate equations with density matrices:
Independent influence of IR on shift and broadening
(P. Lambropoulos et al.)

$$(1)\ \partial_t \sigma_{gg} = -2Im(\Omega_1^* \sigma_{eg})$$
$$(2)\ \partial_t \sigma_{ee} = -(\Gamma_e + \Gamma_{ion})\sigma_{ee} + 2Im(\Omega_1^* \sigma_{eg})$$
$$(3)\ \partial_t \sigma_{eg} = [-i(\Delta_1 - S_e) - 1/2(\Gamma_e + \Gamma_{ion} + \gamma_L)]\sigma_{eg} + i\Omega_1(\sigma_{gg} - \sigma_{ee})$$
$$(4)\ \partial_t \sigma_h = \Gamma_{ion}\sigma_{ee} - \Gamma_h \sigma_h$$
$$(5)\ \partial_t \sigma_f = \Gamma_e \sigma_{ee} + \Gamma_h \sigma_h \equiv \partial_t \sigma_f^R + \partial_t \sigma_f^N$$





Approximation:

Polarizability ~ 1/ω²

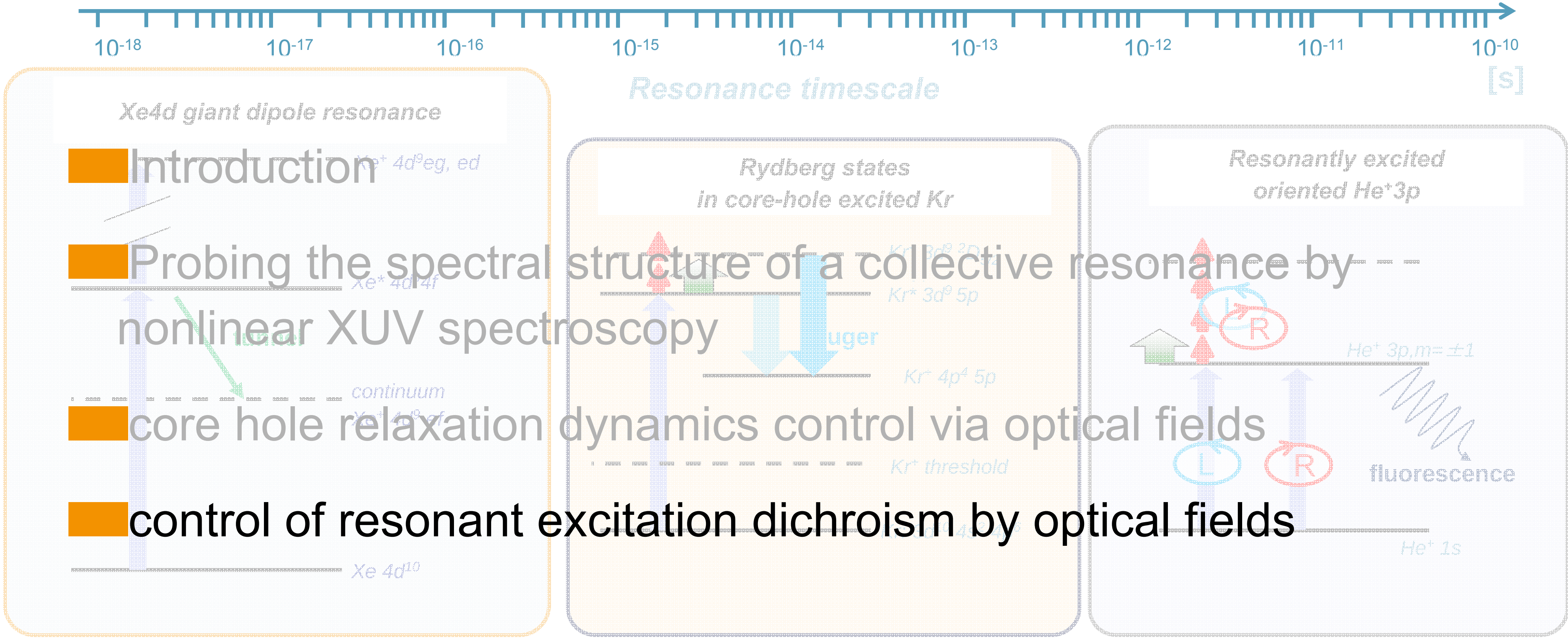
AC Stark shift
~
ponderomotive shift
$U_p = eE_0^2 / 4m\omega^2$

Quasi-free approximation for the Rydberg state:

-No influence of intermediate resonances
-No spin-dependence of polarizability

T. Mazza *et al*, *J. Phys. B* **45** 141001 (2012)

Outline



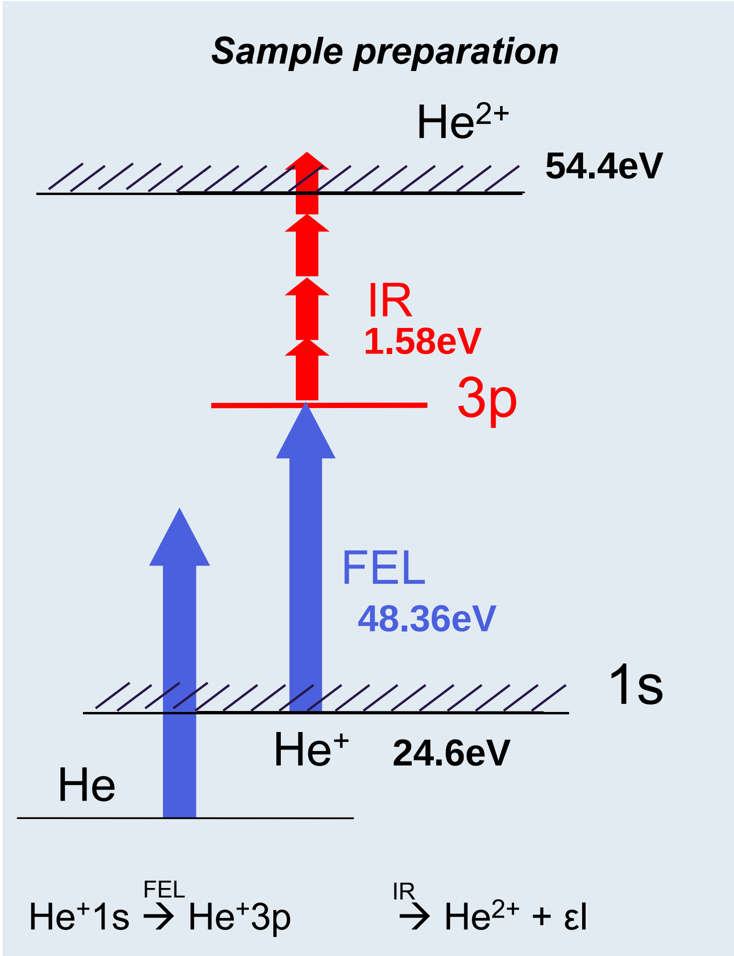
- Introduction
- Probing the spectral structure of a collective resonance by nonlinear XUV spectroscopy
- core hole relaxation dynamics control via optical fields
- control of resonant excitation dichroism by optical fields

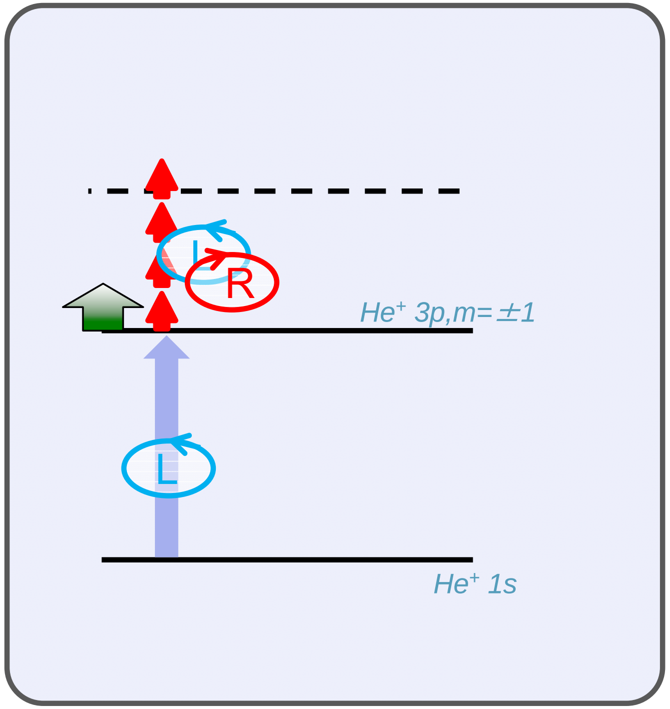
Resonances in intense photon fields: (two-color, circularly polarized light) case 3

1. Dichroism in the photoionization: the magnetic quantum state selectivity of the CIPO light affects significantly the ionization cross section

2. AC stark shift of magnetic quantum number selected electronic state

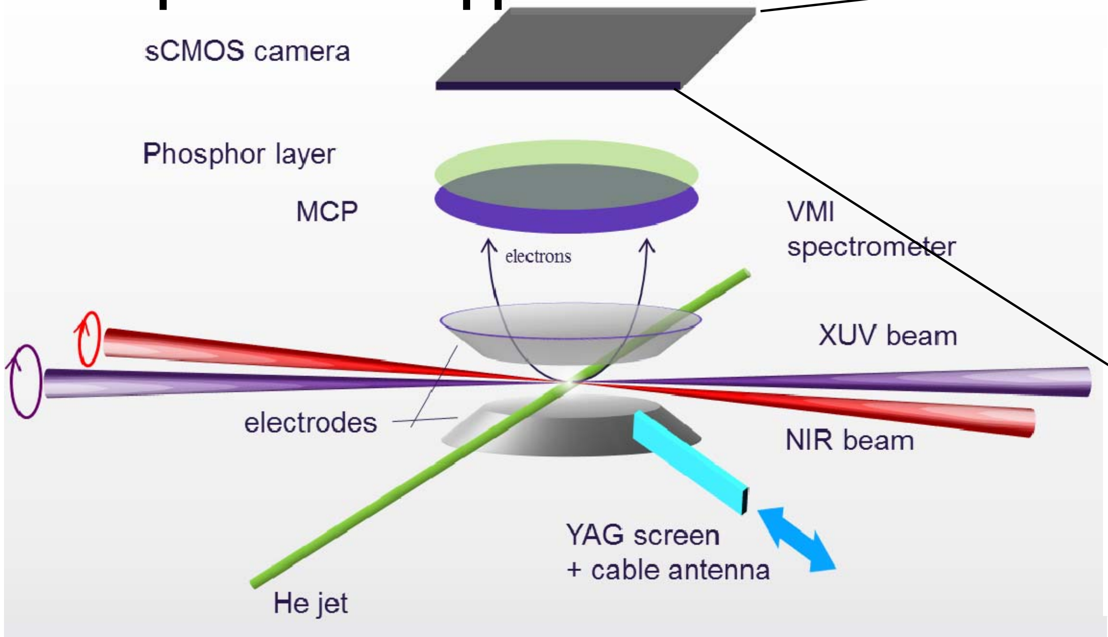
"control of resonant excitation dichroism by intense fields"

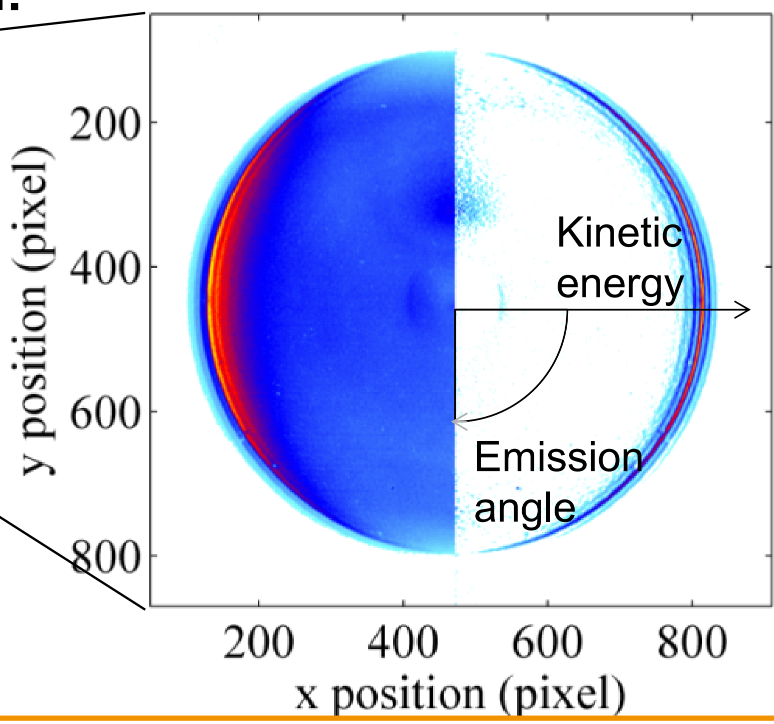




M. Ilchen *et al*, PRL 118, 013002 (2017)

XUV-IR Electron Spectroscopy at LDM@FERMI: Experimental apparatus





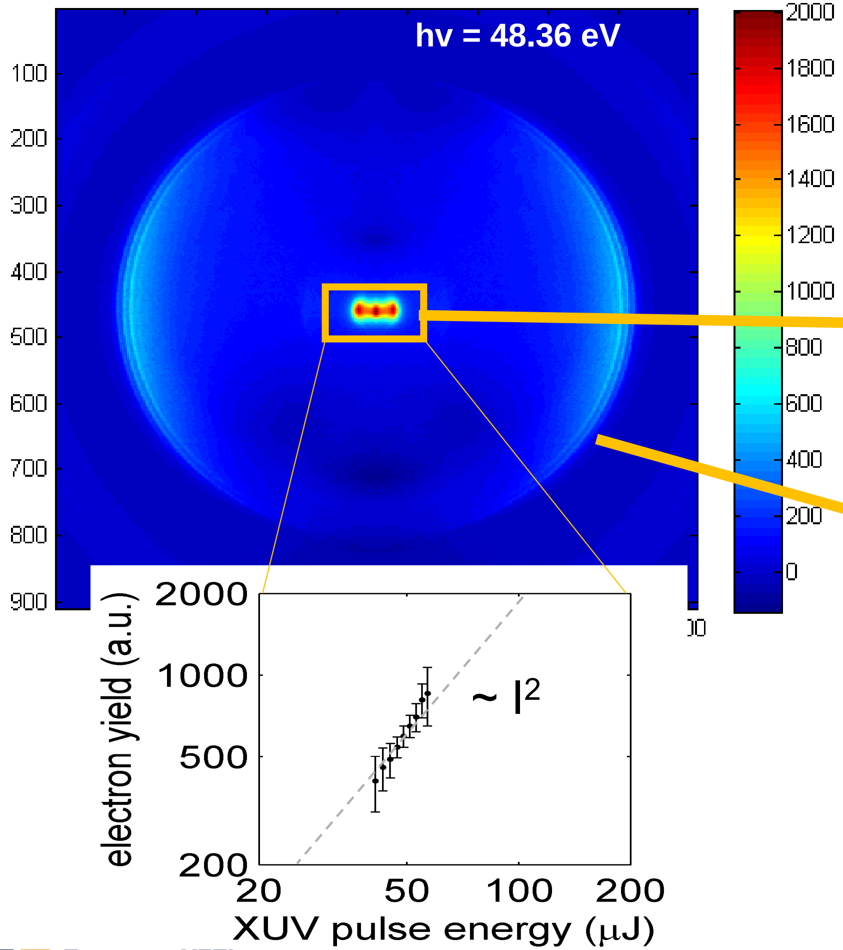
XUV source: FERMI
hv = 48.36 eV (He+1s3p resonance)
BW ~0.1%, **seeded FEL**

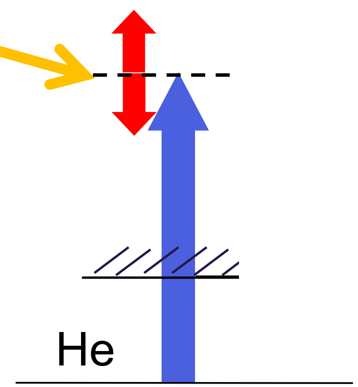
Pulse duration ~ 50-100 fs,
Intensity irrelevant

IR
λ = 800 nm
Intensity ≤ 1.5 10^{12} W/cm^2
No pump probe: time overlap

VMI:
~200meV energy resolution for KE≤4eV
Angular resolution

Electron spectroscopy of the He+3p 2color MPI



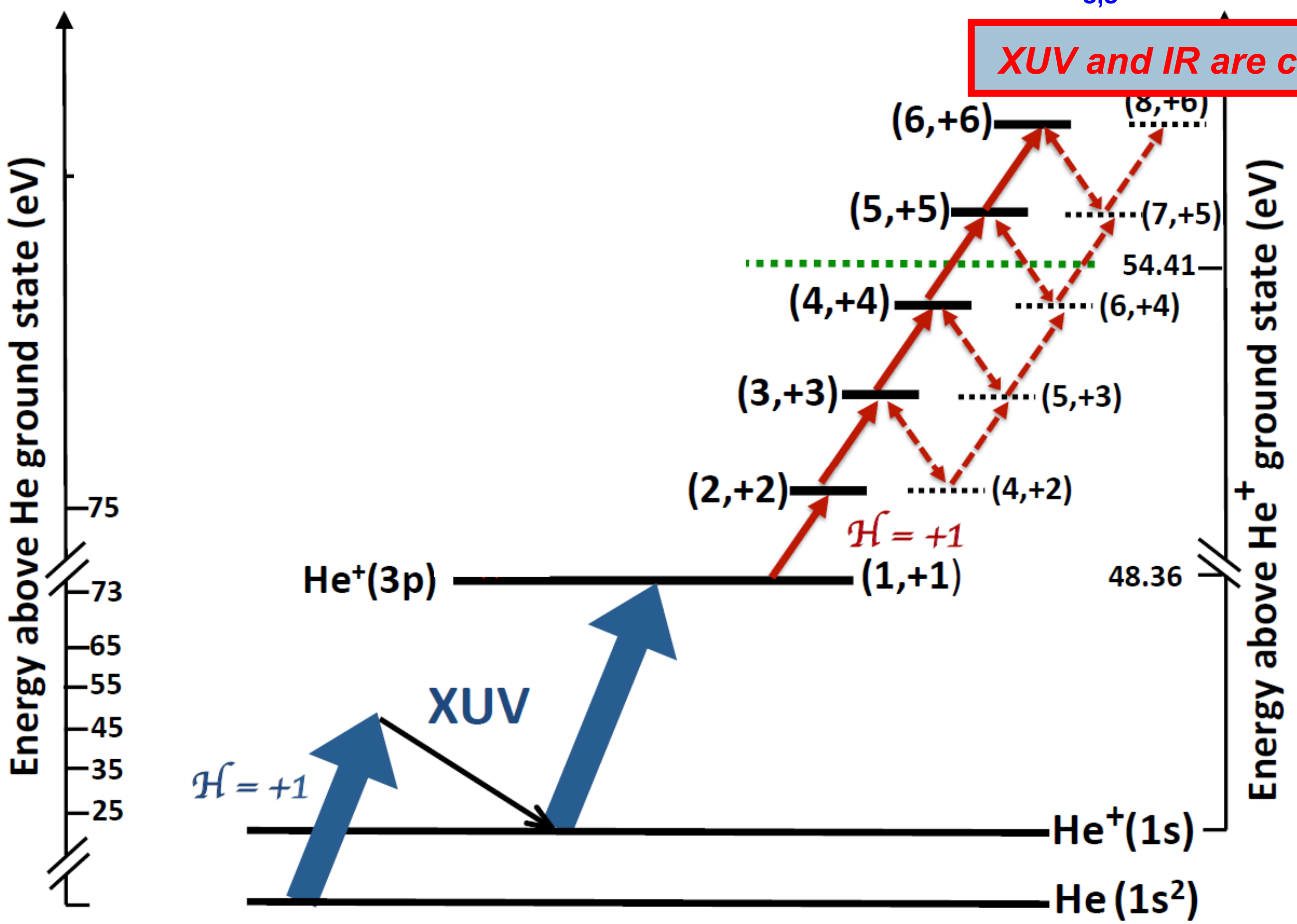


He$^+$1s $\xrightarrow{FEL}$ He$^+$3p $\xrightarrow{IR}$ He^{2+} + εl

Experimental Scheme – NIR Intensity = $0.7*10^{12}$ W*cm^{-2}

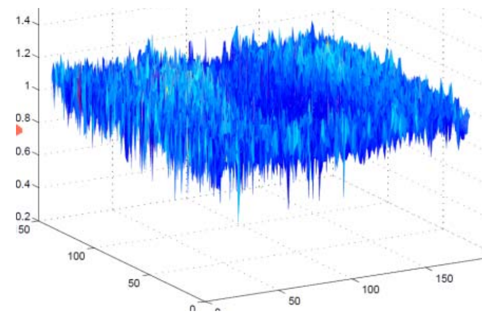
$P_{5,5} = 1.5*10^{-2}$

XUV and IR are co-rotating



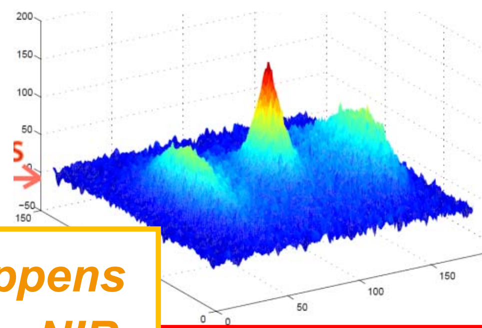
counter-rotating

Full yield range ~ 0.6 (arb. units)



co-rotating

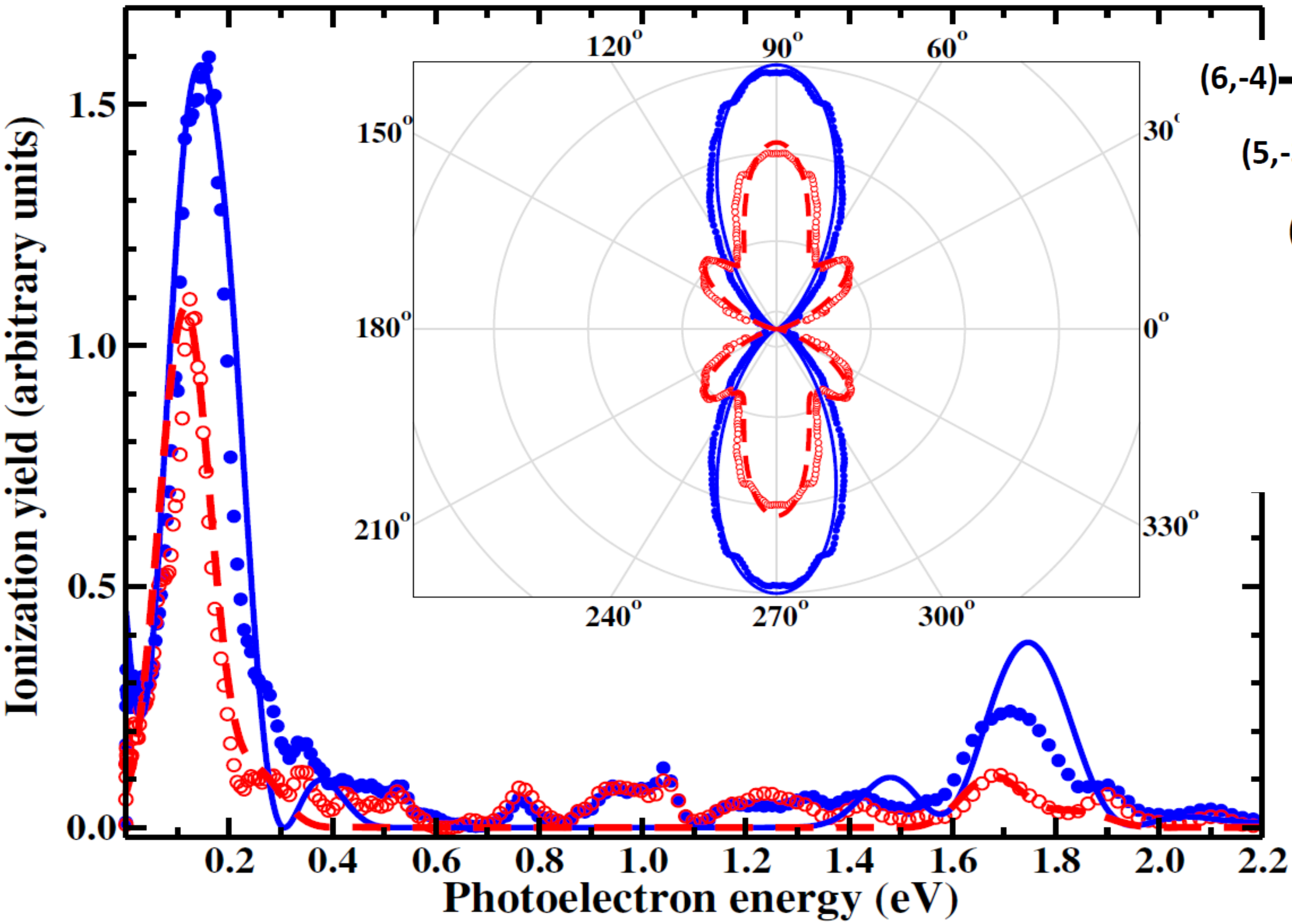
Full yield range ~ 150 (arb. units)



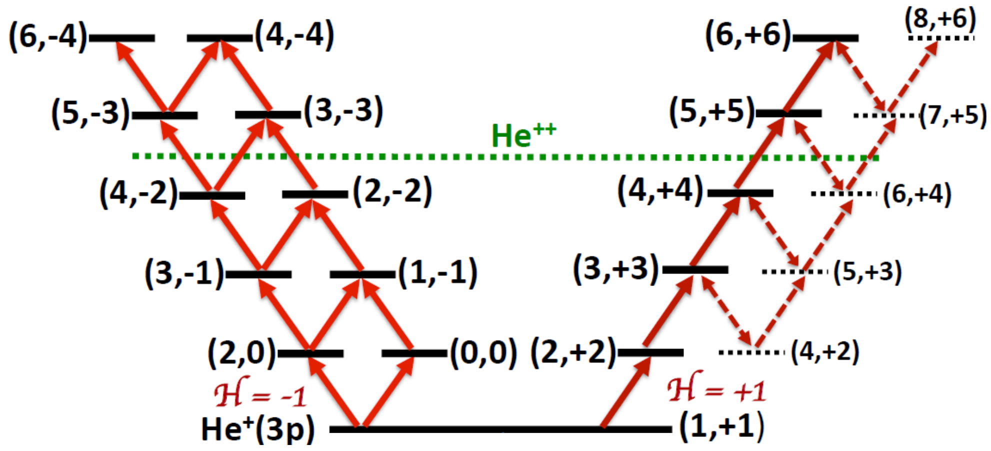
What happens at higher NIR intensity?

M. Ilchen *et al*, PRL 118, 013002 (2017)

Photoelectron Circular Dichroism – Yield



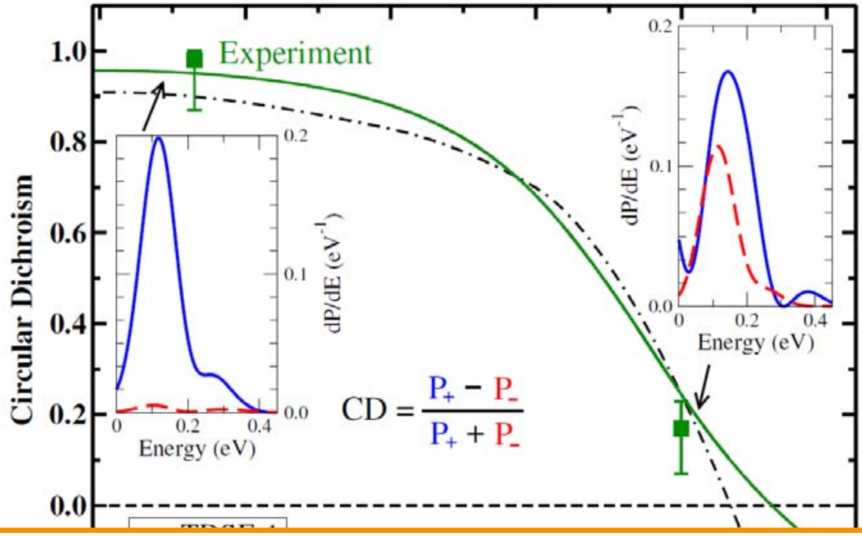
NIR Intensity = $1.4*10^{12}$ W/cm²



- Evidence of 1 partial wave vs. 2 partial wave contribution in the PAD
- Intensity increase x2 → yield increase $x2^4$ = 16??

M. Ilchen *et al*, PRL 118, 013002 (2017)

Photoelectron Circular Dichroism – Intensity Dependence

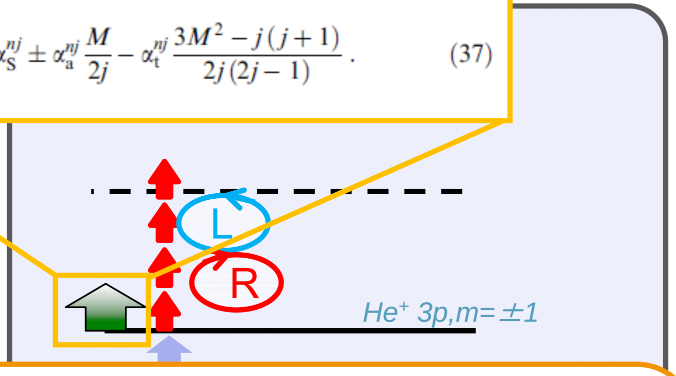


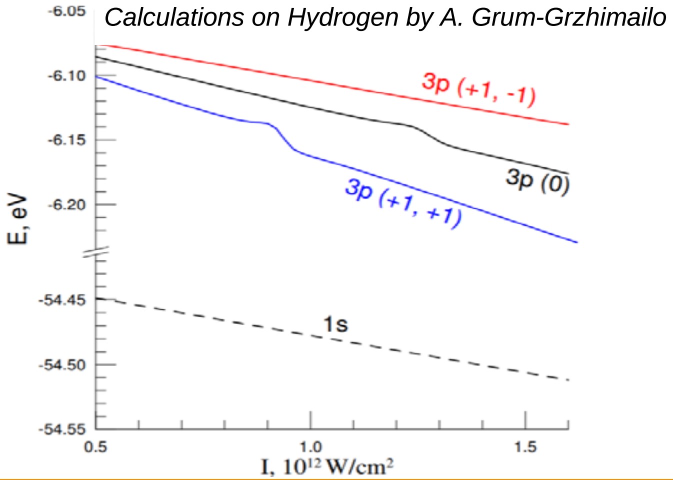
TDSE1 by Kabachnik et al.
TDSE2 by Bartschat et al.

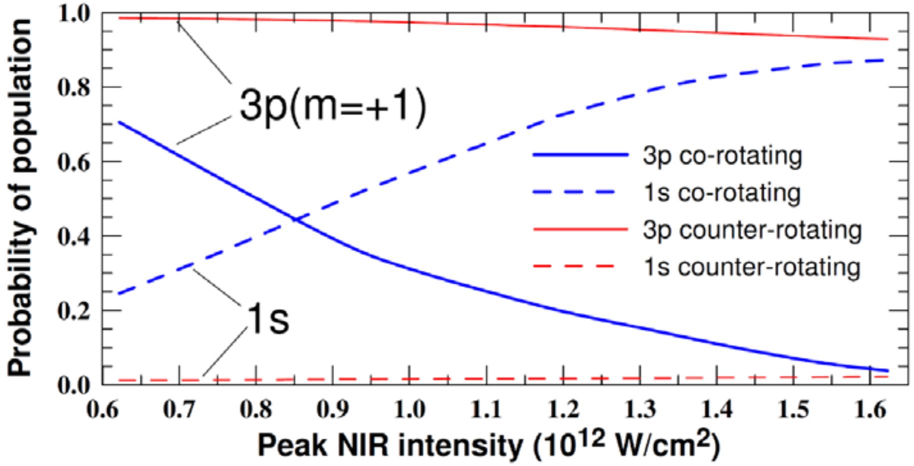
Delone and Krainov, Physics - Uspekhi 42 (7) 669 - 687 (1999)

In the case of circular polarization, the use of the Wigner–Eckart theorem leads to the following explicit dependence of the dynamic polarizability on the magnetic quantum number M:

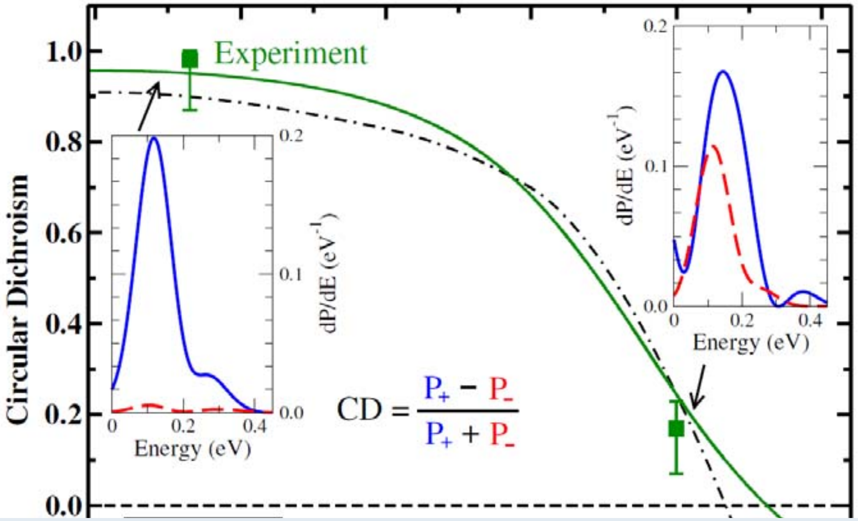
$$\alpha^{njM}(\omega) = \alpha_S^{nj} \pm \alpha_a^{nj}\frac{M}{2j} - \alpha_t^{nj}\frac{3M^2 - j(j+1)}{2j(2j-1)}. \quad (37)$$







Dichroic Stark shift



Delone and Krainov, Physics - Uspekhi 42 (7) 669 - 687 (1999)

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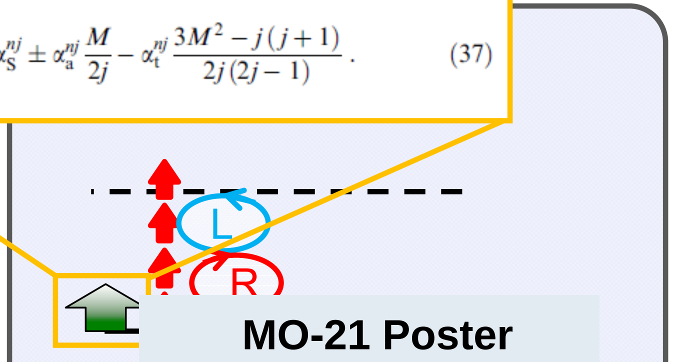
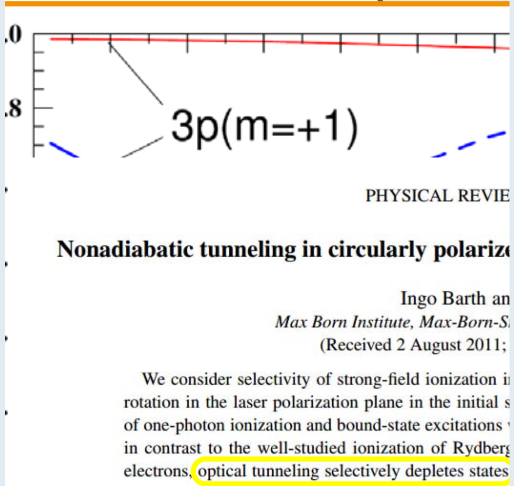
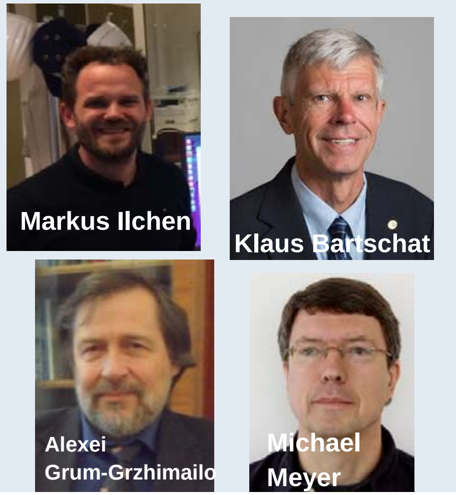


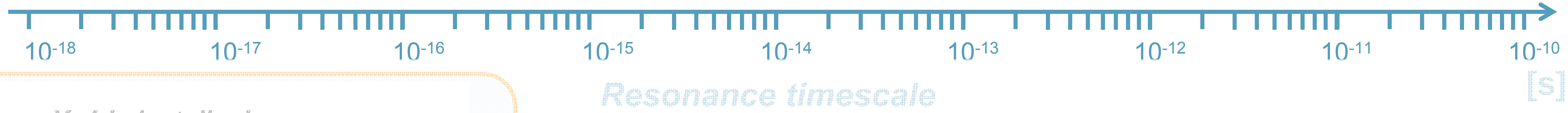
- Electron population of the He⁺ 3p (m=+1) state is strongly NIR-intensity dependent because of dichroich Stark shift
- This effect partially is in competition with the co-rotating ionization cross section (due to angular factors alone)
- A combination of the population control and together with the expected sign change of the CD points to unexpectedly low intensity for a sign change (compared to e.g. Barth and Smirnova. (2011), Bauer et al. (2014)).



MO-21 Poster



Conclusions



- Probing the spectral structure of a collective resonance by nonlinear XUV spectroscopy
  - Substructure in the Xe4d GDR unveiled by 2photon ATI spectroscopy
- Core hole relaxation dynamics control via optical fields
  - Dynamic Stark shift of core hole resonance, introducing a competition between the optical ionization and the Auger decay
- Control of resonant excitation dichroism by optical fields
  - Intensity dependent dichroism evidencing the influence of the magnetic state on the optical modification of the excitation process