Dichroism and resonances in intense radiation fields

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Pics:
www.xfel.eu
www.desy.de
maps.google.com
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**Experiment**

**Theory**

http://static.naukas.com/media/2012/05/8-Winkel-tripel-projection.jpg
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**

- $10^{-18}$ to $10^{-10}$

**Resonance timescale**

**Rydberg states in core-hole excited Kr**

- $Kr^+$ $3d^9 \, 2D_{5/2}$
- $Kr^+$ $3d^9 \, 5p$
- $Kr^+$ $4p^4 \, 5p$
- $Kr^+$ threshold
- $Kr: 3d^{10} \, 4s^2 \, 4p^6$

**Xe4d giant dipole resonance**

- $Xe^+ 4d^24f$
- tunnel
- continuum $Xe^+ 4d^9 ef$

**Resonantly excited oriented He+3p**

- $He^+ 1s$
- $He^+ 3p, m = \pm 1$
- fluorescence

**European XFEL**
**Dichroism and resonances in intense radiation fields**

- **Resonance timescale**

- **Rydberg states in core-hole excited Kr**
  - Kr$^+$ 3d$^9$ 2D$_{5/2}$
  - Kr$^+$ 3d$^9$ 5p
  - Kr$^+$ threshold
  - Kr: 3d$^{10}$ 4s$^2$ 4p$^6$

- **Xe4d giant dipole resonance**
  - Xe$^+$ 4d$^9$ eg, ed
  - Xe$^+$ 4d$^9$4f
  - continuum Xe$^+$ 4d$^9$ ef
  - Xe 4d$^{10}$

- **Resonantly excited oriented He$^+$3p**
  - He$^+$ 4d giant dipole resonance
  - He$^+$ 1s
  - He$^+$ 3p,$m=±1$
  - fluorescence
Outline

- 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
Outline

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

- 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:

\[
\hat{H} = \sum_{n=1}^{N} \left( \frac{\hat{p}_n^2}{2} - \frac{Ze}{|\hat{r}_n|} \right) + \frac{1}{2} \sum_{n,n' = 1}^{N} \sum_{n \neq n'}^{N} \frac{\delta \left\langle \frac{1}{2} \right\rangle}{|\hat{r}_n - \hat{r}_{n'}|} =: \hat{H}_0 + \hat{H}_1,
\]

- **“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 \textit{ab initio} theory seen (understood) by an experimentalist

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

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

\[
\sigma^{(2)} = \left| \sum_{M_{\text{res}}} \frac{\langle F | \hat{H}_{\text{int}} | M_{\text{res}} \rangle \langle M_{\text{res}} | \hat{H}_{\text{int}} | I \rangle}{E - E_{M_{\text{res}}} + \frac{i}{\Gamma_{\text{res}}}} \right|
\]

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

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

<table>
<thead>
<tr>
<th>( \Xi_n ) (eV)</th>
<th>( \Gamma_n ) (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Intrachannel</td>
<td></td>
</tr>
<tr>
<td>4d_0</td>
<td>76.3</td>
</tr>
<tr>
<td>4d_{\pm 1}</td>
<td>77.6</td>
</tr>
<tr>
<td>4d_{\pm 2}</td>
<td>77.2</td>
</tr>
<tr>
<td>Full CIS</td>
<td></td>
</tr>
<tr>
<td>( R_1 )</td>
<td>74.3</td>
</tr>
<tr>
<td>( R_2 )</td>
<td>107.2</td>
</tr>
</tbody>
</table>

\( ^a \) All SES values have an error bar of 0.1 eV. This is ca Chen et al., PRA 91, 032503 (2015)
**XUV non-linear Spectroscopy at FLASH: Experimental apparatus**

**MBES:** $4\pi$ collection angle
- Suited to highly dilute samples
- Enabling single shot capability

**XUV source:** FLASH
$hv = 105, 140 \text{ eV}$
$BW \sim < 1 \text{ eV}, \text{ SASE process}$

Pulse duration $\sim 50-100 \text{ fs}$,
focus $\sim 3-5 \text{ um}, \text{ gaussian model}$

$10 \text{ uJ} \sim 10^{14} - 10^{15} \text{ W/cm}^2$

**MBES:**
$\Delta KE / KE \sim 2\%$ on ret. electrons
Single shot capability ($4\pi$ acceptance)

**Sample delivery:**
Sample density can be tuned over $\sim 2 \text{ o.m.}$ in a controlled way
Above Threshold Ionization of Xe4d at hv = 105eV
Above Threshold Ionization of Xe4d at $h\nu = 105\text{eV}, 140\text{eV}$: 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

Interchannel coupling

Intrachannel coupling

Interchannel vs. intra $\sigma(\omega)$

- blue-shift
- broadening

T. Mazza et al., Nature Comms. 6, 6799 (2015)
Dichroism and resonances in intense radiation fields

Tomaso Mazza. SQS Instrument Scientist, ICPEAC30 – July 31th 2017

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

Interchannel coupling

The blue curve is the black (1ph) curve times a $E^{-13/2}$ free-free transition probability

Intra $\sigma(\omega)$ vs. $\sigma^{(2)}(\omega)$

- red-shift and sharpening
- Consistent with 2-factorization

Inter $\sigma(\omega)$ 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”

S. E. Harris, Physics Today 50, 7, 36 (1997)
XUV-IR Electron Spectroscopy at FLASH: Experimental apparatus

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

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

Pulse duration ~ 50-100 fs,
Intensity irrelevant

**IR:**
- λ = 800 nm
- Intensity ~ 10^{12} W/cm²

**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

![Diagram showing transitions and energy levels](image)

Kr* 3d\(^9\)2D\(_{5/2}\)

Kr* 3d\(^9\)5p

XUV: 91.2 eV

Kr: 3d\(^{10}\) 4s\(^2\) 4p\(^6\)

hv = 91.2 eV
Electron Spectroscopy:
Auger decay suppressed by dressing IR field

\[ S = f(NIR) \]

Kr: 3d^{10} \, 4s^2 \, 4p^6

Kr\(^+\): 3d^{9}\, 4s^2\, 4p^5

XUV & NIR

\[ \text{hv} = 91.2 \text{ eV} \]
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

\[ I_{IR} = 2.3 \cdot 10^{11} \text{ W/cm}^2 \]
\[ \Delta E = 15 \pm 5 \text{ meV} \]
\[ W_{\text{laser on}} = 160 \pm 15 \text{ meV} \]

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

$I_{IR} = 4.4 \cdot 10^{11} \text{ W/cm}^2$

$\Delta E = 28 \pm 15 \text{ meV}$

$W_{\text{laser on}} = 255 \pm 40 \text{ meV}$

Dichroism and resonances in intense radiation fields

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

\[ I_{IR} = 8.9 \times 10^{11} \text{ W/cm}^2 \]
\[ \Delta E = 39 \pm 17 \text{ meV} \]
\[ W_{\text{laser on}} = 287 \pm 54 \text{ meV} \]

Dichroism and resonances in intense radiation fields


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

\[ I_{IR} = 1.9 \cdot 10^{12} \text{ W/cm}^2 \]

\[ \Delta E = 97 \pm 29 \text{ meV} \]

\[ W_{\text{laser on}} = 506 \pm 110 \text{ meV} \]
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.)

\[
\begin{align*}
1. \quad \partial_t \sigma_{gg} &= -2i \Omega_1 \sigma_{eg} \\
2. \quad \partial_t \sigma_{ee} &= -(\Gamma_e + \Gamma_{ion}) \sigma_{ee} + 2i \Omega_1 \sigma_{eg} \\
3. \quad \partial_t \sigma_{eg} &= [i(\Delta_1 - S_e) - 1/2(\Gamma_e + \Gamma_{ion} + \gamma_f)] \sigma_{eg} \\
&\quad + \Omega_1 (\sigma_{gg} - \sigma_{ee}) \\
4. \quad \partial_t \sigma_{h} &= \Gamma_{ion} \sigma_{ee} - \Gamma_h \sigma_h \\
5. \quad \partial_t \sigma_{f} &= \Gamma_e \sigma_{ee} + \Gamma_h \sigma_h \equiv \partial_t \sigma_{f}^R + \partial_t \sigma_{f}^I
\end{align*}
\]

\[S = U_p = eE_0^2 / 4m \omega^2\]

Approximation:
\[\text{Polarizability} \sim 1/\omega^2\]

\[\text{AC Stark shift} \sim \text{ponderomotive shift}\]
\[U_p = eE_0^2 / 4m \omega^2\]

Quasi-free approximation for the Rydberg state:
- No influence of intermediate resonances
- No spin-dependence of polarizability

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- 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

Rydberg states in core-hole excited Kr

- Kr: 3d\textsuperscript{10} 4s\textsuperscript{2} 4p\textsuperscript{6}
- Kr\textsuperscript{+}: 3d\textsuperscript{9}\textsuperscript{2} D\textsuperscript{5/2}
- Kr\textsuperscript{*}: 3d\textsuperscript{9} 5p

Auger continuum

Xe: 4d\textsuperscript{10}

Xe\textsuperscript{+}: 4d\textsuperscript{9} eg, ed

Xe\textsuperscript{+}: 4d\textsuperscript{9} 5f

He\textsuperscript{+}: 1s

He\textsuperscript{+}: 3p, m=±1

Xe\textsuperscript{4d} giant dipole resonance

Resonance timescale

- 10\textsuperscript{-18} - 10\textsuperscript{-14}
- 10\textsuperscript{-15} - 10\textsuperscript{-11}
- 10\textsuperscript{-12} - 10\textsuperscript{-10}

Xe\textsuperscript{4d} giant dipole resonance

European XFEL
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)
Dichroism and resonances in intense radiation fields

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Sample: Atoms, molecules, clusters (including heterogeneous / doped / metal clusters) in supersonic beams


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

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

Electron yield (a.u.)

XUV pulse energy (μJ)

hv = 48.36 eV

He

He^+1s → He^+3p

FEL

IR

→ He^{2+} + εl
Dichroism and resonances in intense radiation fields

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

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

$P_{5,-3} = 2.7 \times 10^{-4}$

$P_{5,5} = 2.3 \times 10^{-4}$

Full yield range ~ 0.6 (arb. units)

Full yield range ~ 150 (arb. units)

What happens at higher NIR intensity?

M. Ilchen et al, PRL 118, 013002 (2017)
NIR Intensity $= 1.4 \times 10^{12}$ W/cm$^2$

Evidence of 1 partial wave vs. 2 partial wave contribution in the PAD

Intensity increase $\times 2 \rightarrow$ yield increase $\times 2^4 = 16$?

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 dichroism, the use of the Wigner–Eckart theorem leads to the following explicit dependence of the dynamic polarizability on the magnetic quantum number $M$:

$$\chi^{\mu M}(\omega) = \chi_0^{\mu M} \pm \frac{M}{2j} \left[ 3M^2 - j(j+1) \right] \frac{1}{2j(2j-1)}.$$  (37)

Calculations on Hydrogen by A. Grum-Grzhimailo
**Dichroic Stark shift**

- Electron population of the He$^+$ 3p ($m=+1$) state is strongly NIR-intensity dependent because of dichroic 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)).
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