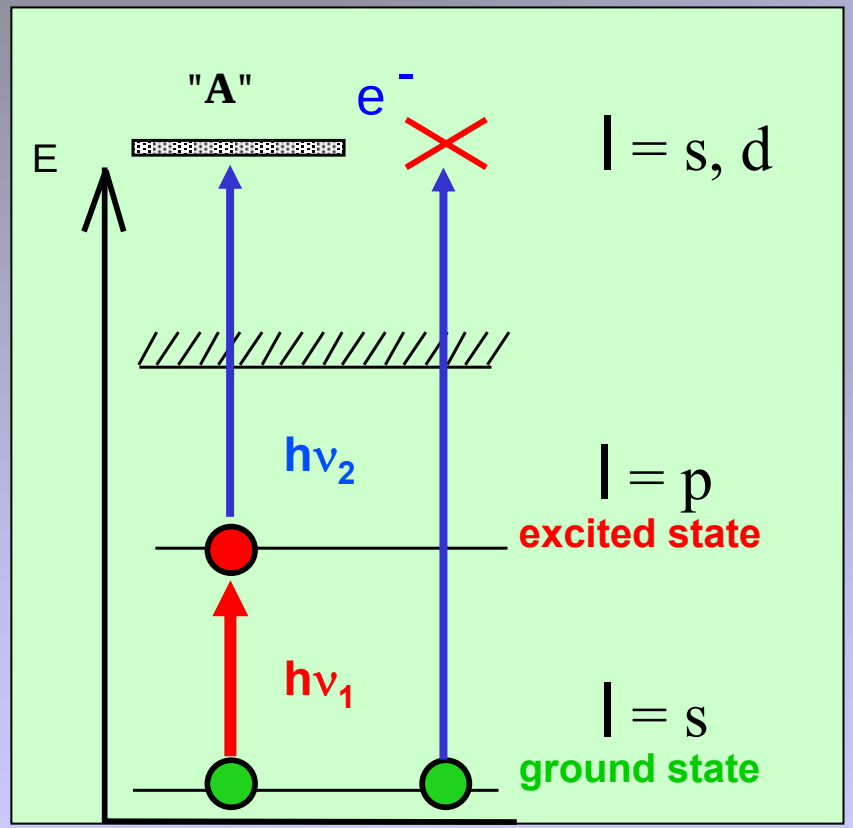
Pump-Probe Experiments in the Gas Phase using the TESLA-FEL

Michael Meyer, L.U.R.E., Orsay, France

- Introduction
  - **Pump-Probe Experiments**
  - **SR + opt. Laser <--> FEL + opt. Laser**
- FEL characterization
  - **Cross correlation experiments on atomic Ar**
- Electronic correlations in excited atomic states
  - **Coupling of autoionization states**
- Dynamics of molecular fragmentation
  - **Time-resolved analysis**
  - **Quantum beat spectroscopy**
- Conclusion

Pump - Probe - Experiments
photoexcited states (atomic case)



electron interaction

Preparation of initial state

- **manipulation of electron cloud**
- **controlled change of interaction**

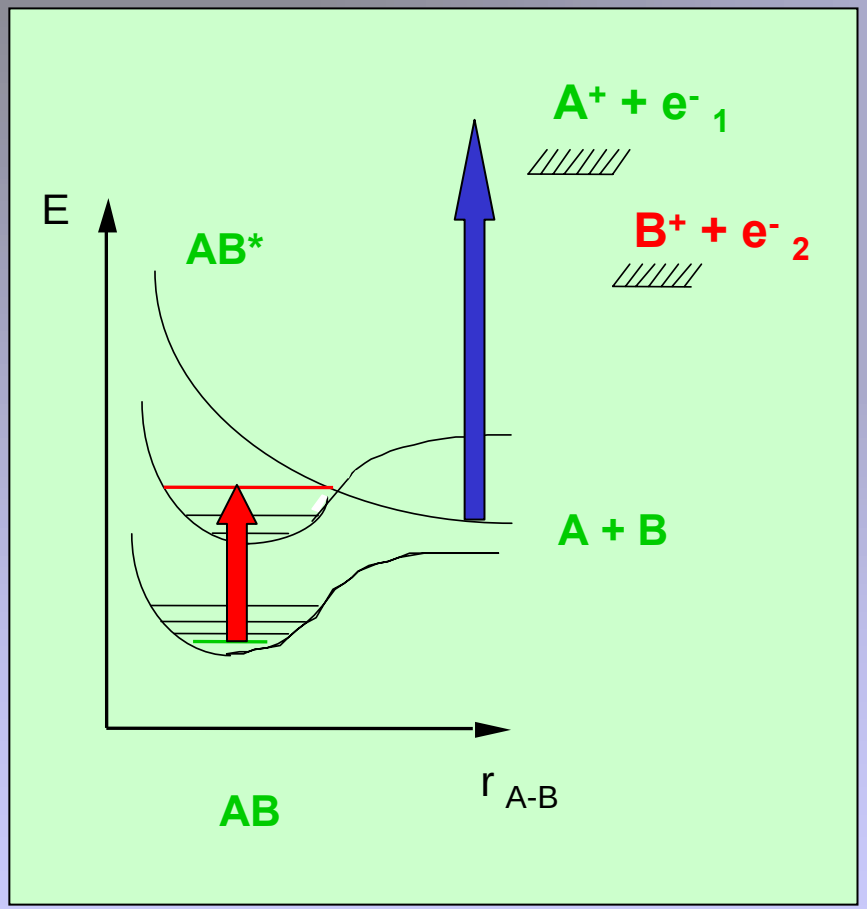
→ **Probe by photoionization**

"Selection" of final state

- **two-photon excitation**
- **partial cross sections**

comparison with theoretical models

Pump - Probe - Experiments
photoexcited states (molecular case)



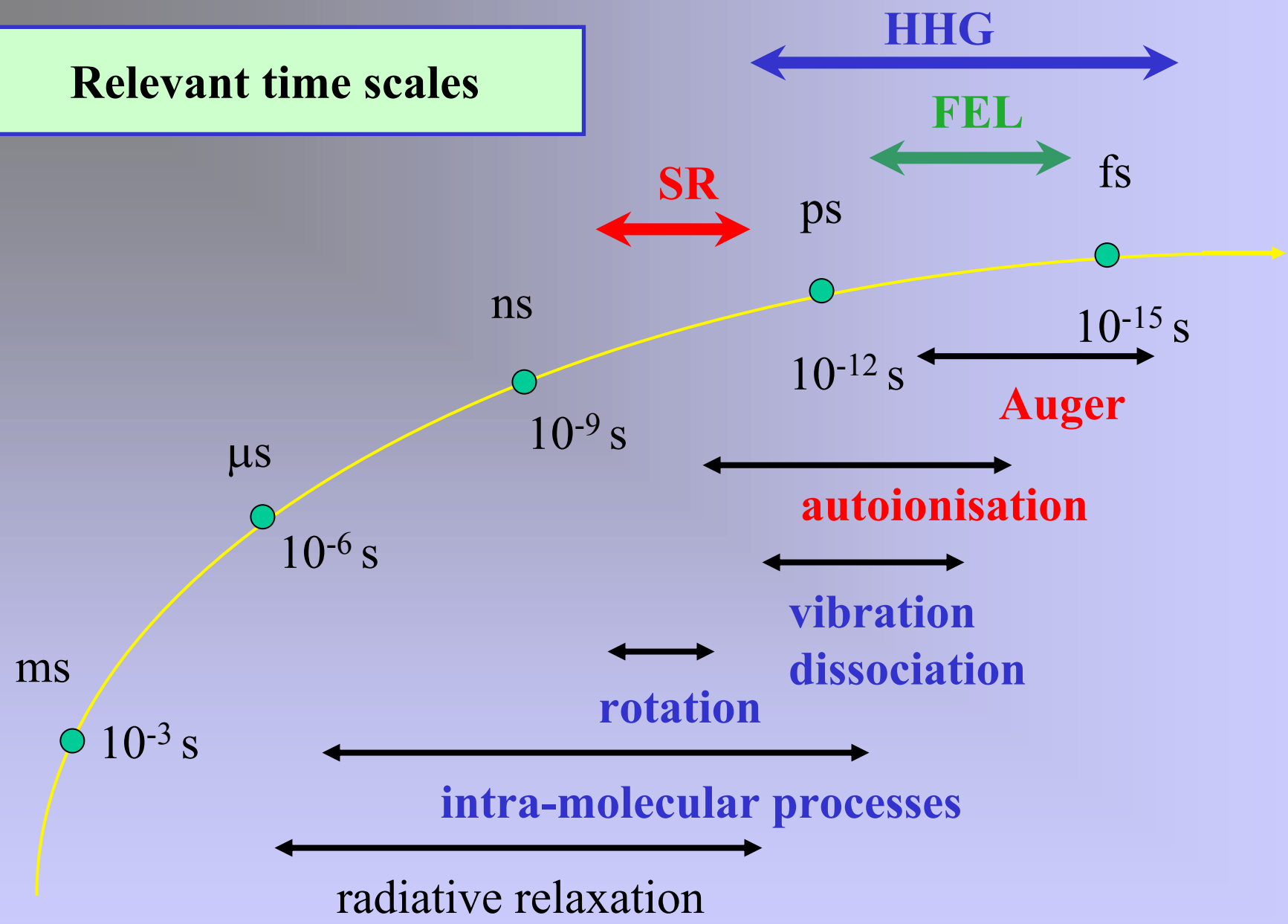
photodissociation

- **photoinduced processes**
- **characterization of products**
- **dynamics of excited states**

---> pulsed sources

time-evolution of the process

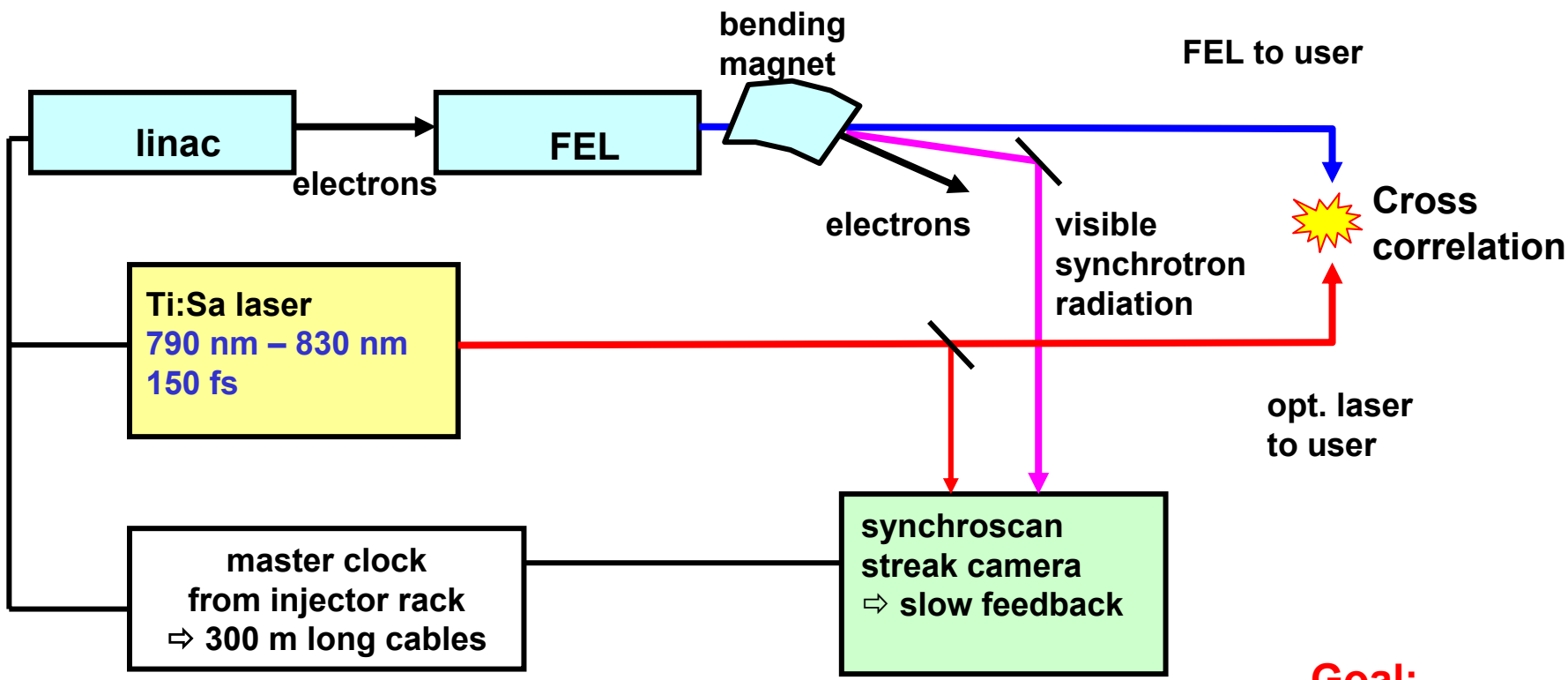
Relevant time scales



Characteristics of laser and synchrotron sources

	laser	SR	FEL
$\Delta E / E$	10^{-9}	10^{-5}	10^{-3}
ph/s ph/pulse	10^{18} 10^{11}	10^{12} 10^{5}	10^{15} 10^{14}
$h\nu$	IR - UV (1μm - 300nm)	IR - X-Ray (≈1eV - keV)	VUV -XUV 15 - 200eV
tuneability	limited	easy	limited
rep. rate	c.w. or pulsed 8.32 MHz	pulsed 8.32 MHz	pulsed 10 Hz
pulse width	Ar^+ : 250 ps dye : <10 ps	600ps ---> 50ps	50 - 200 fs

Synchronization FEL - optical Laser

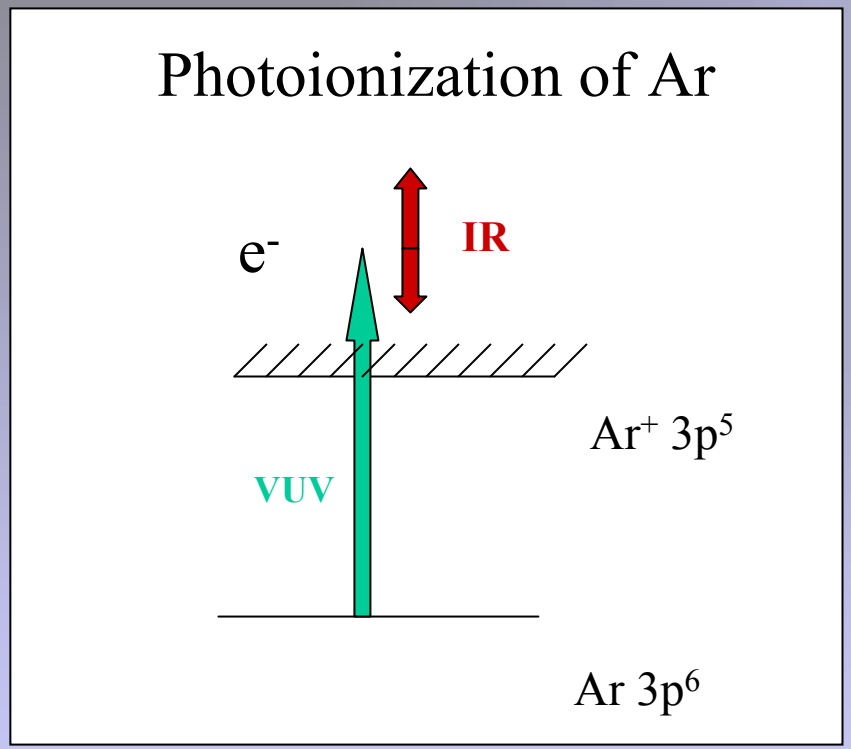


Goal: synchonization < 200 fs

- HASYLAB/DESY (Coordinator)
- Research Centre Jülich / BESSY
- Max-Born-Institute, Berlin
- Dublin City University
- CNRS / LURE
- Lund Laser Centre / MAX-Lab

Funded by the 5th Framework Programme of the European Commission

Characterization of Pump-Probe time-structure and stability using cross correlation experiments



presence of IR:

- shift of IP
- broadening of PES peaks
- sidebands

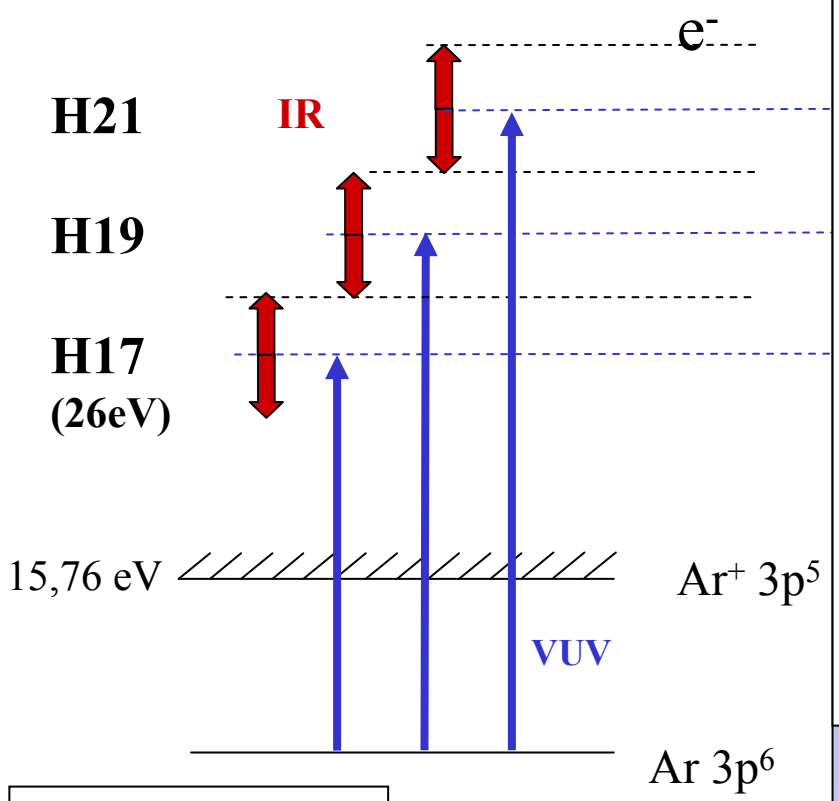
----> $\Delta t(max) = 3fs$

Ti:Saph: 800nm, 40fs, 1 kHz
dressing beam: up to 50µJ --> 13 TW/cm^2

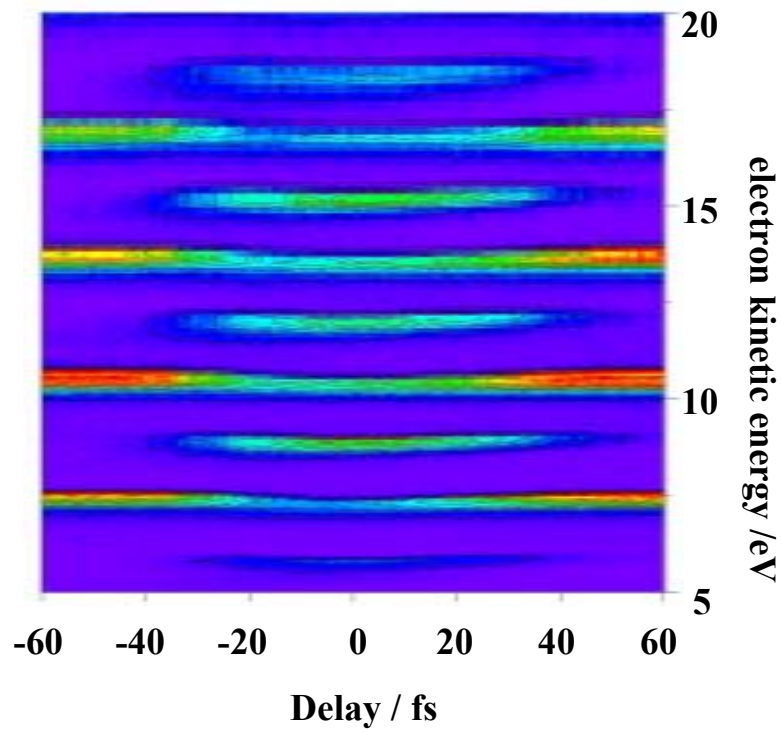
E.S. Toma, H.G. Muller, P.M. Paul, P. Berger, M. Cheret, P. Agostini, C. LeBlanc, G. Mullot, G. Cheriaux
Phys. Rev. A 62, 061801 (2000)

Two-photon (femtosecond)photoionization of Ar

XUV-IR Cross-Correlation



ΔT (laser) = 30fs

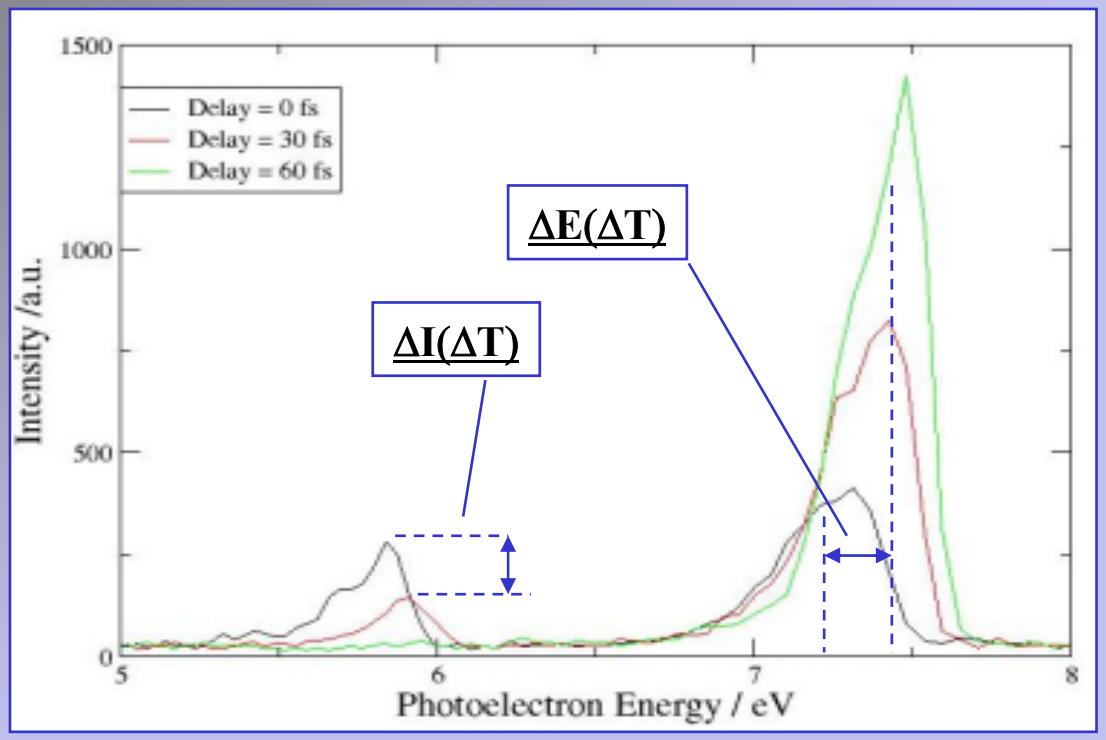


Low field regime: Int(IR) ≈ 10^{11} W/cm^2

$\Gamma(\text{sideband})^2 = \Gamma(\text{IR})^2 + \Gamma(\text{VUV})^2$

P.O'Keeffe, M. Meyer (LURE), V. Veniard, R. Taïeb, A. Maquet (LCP-MR), R. Lopez-Martens, J. Mauritsson, A. Johannson, R. Lopez-Martens, A. L'Huillier (LLC, Lund), PRA69, 051401(R) (2004)

Cross correlation experiments using high order harmonics

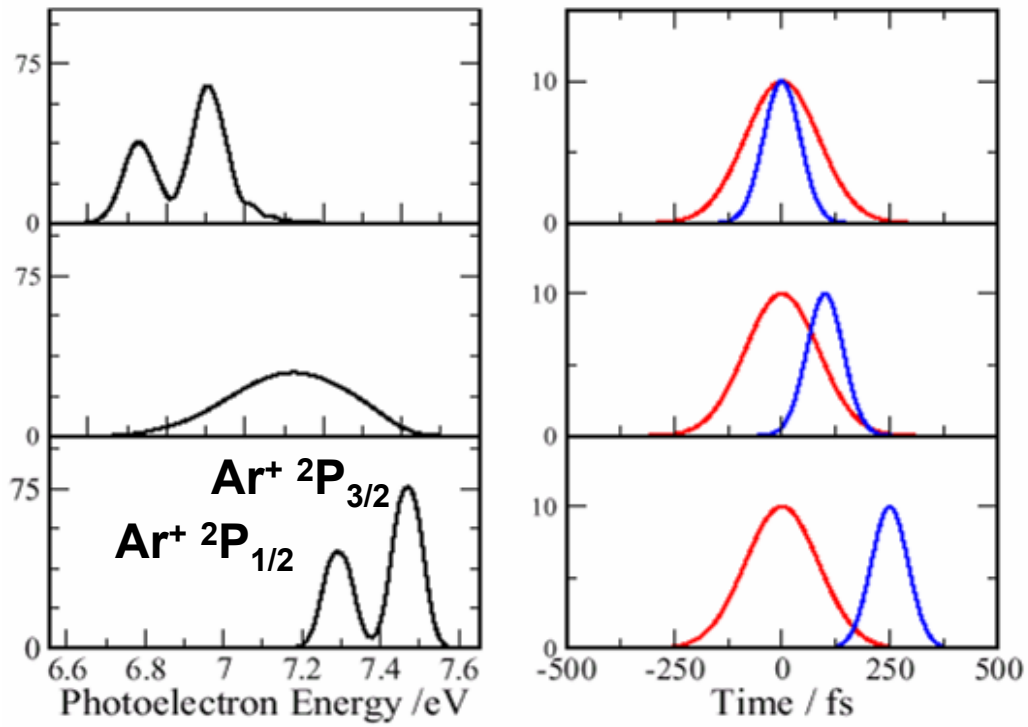


$$I_{eff} \sim \Delta T \text{ ------> } \Delta E\,(\Delta T)$$

$$I_{SB}\,(\Delta T) \sim \int_{-\infty}^{+\infty} f\,[\,I_{IR}(\,t - \Delta T)]\; I_{VUV}(t)\; dt$$

High Intensity Regime: Effect of the ponderomotive shift on the harmonic peak

Photoelectron IR and XUV pulses



I_{IR} (max) = 10 TW/cm^2

τ_{IR} = 200 fs

τ_{XUV} = 100 fs

Resolution E_{kin} = 125 meV

LLC - experiment (HHG):

I (VUV) ≈ 10^6 - 10^7 ph. / pulse

Single pulse : ~ 3 e^- / pulse

---> Σ 30 000 shots

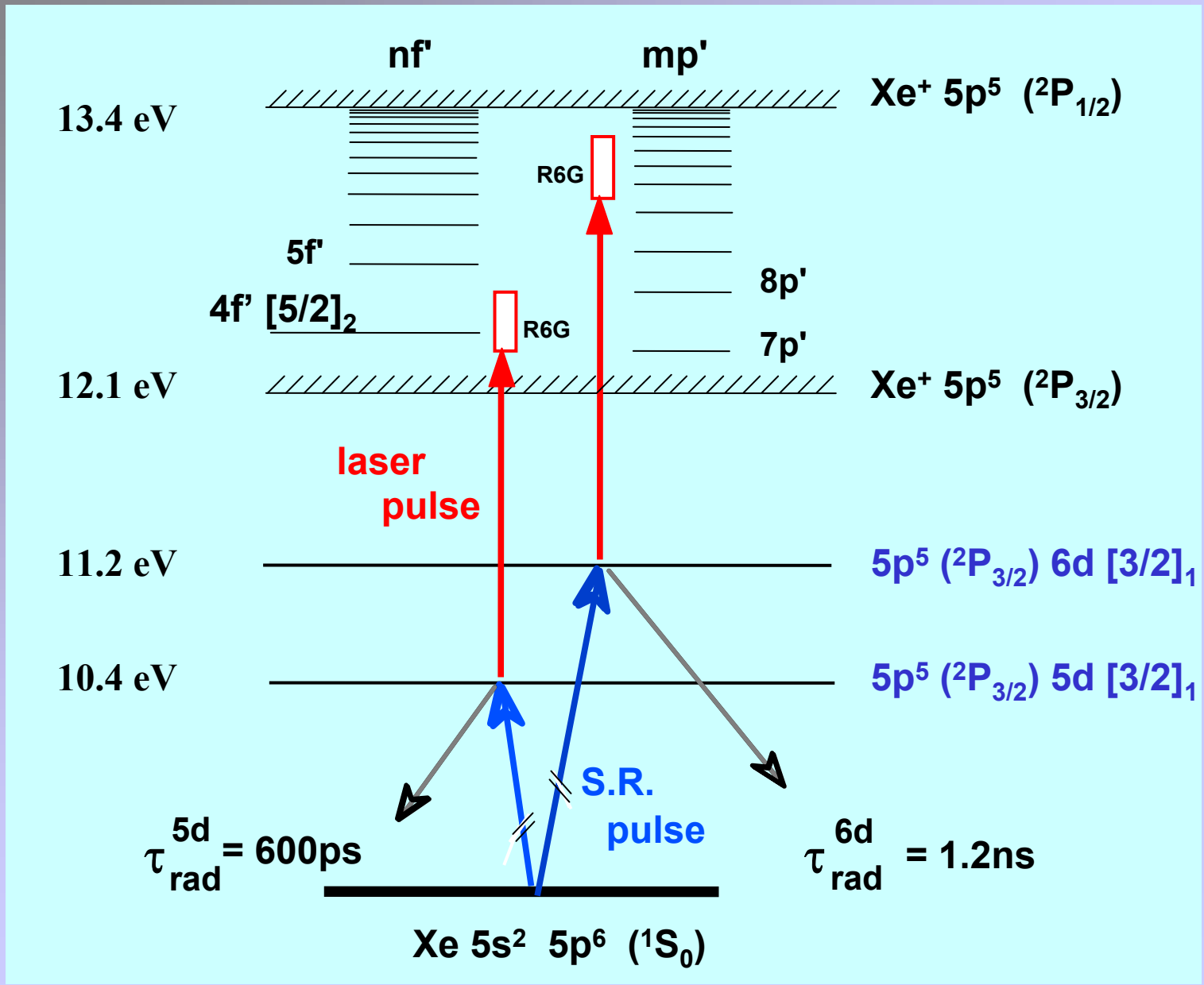
I (FEL) ≈ 10^{13} ph. / pulse

→ Single shot experiments

Application:

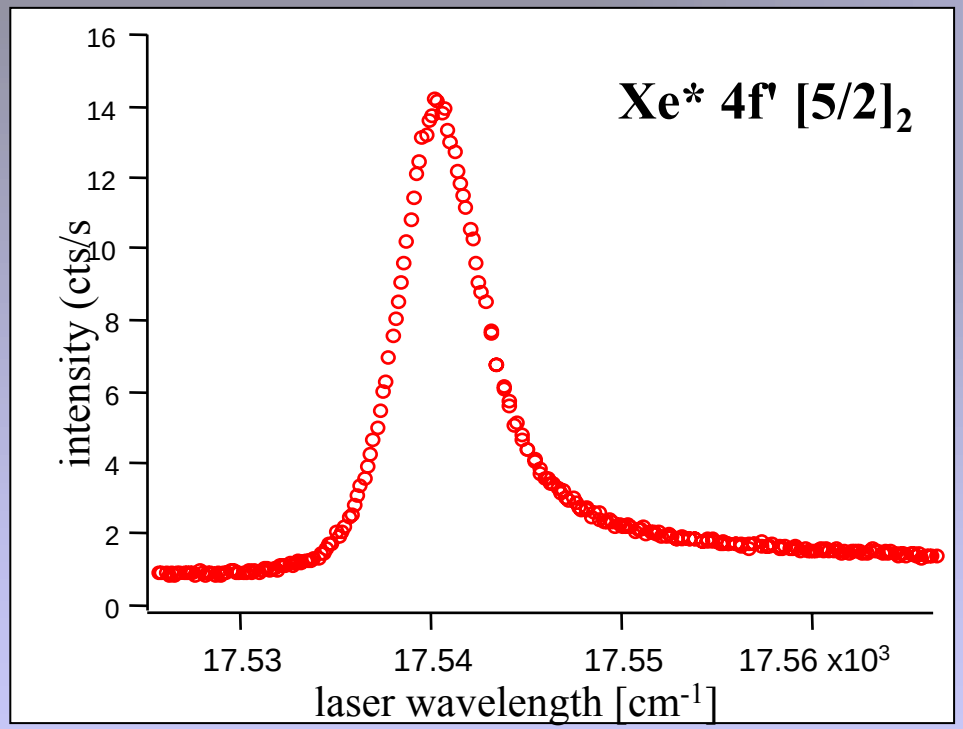
Electronic Correlation in Excited Atomic States

Two-photon double-resonant ionization (SR + Laser)



Two-photon excitation of even-parity states (SR + Laser)

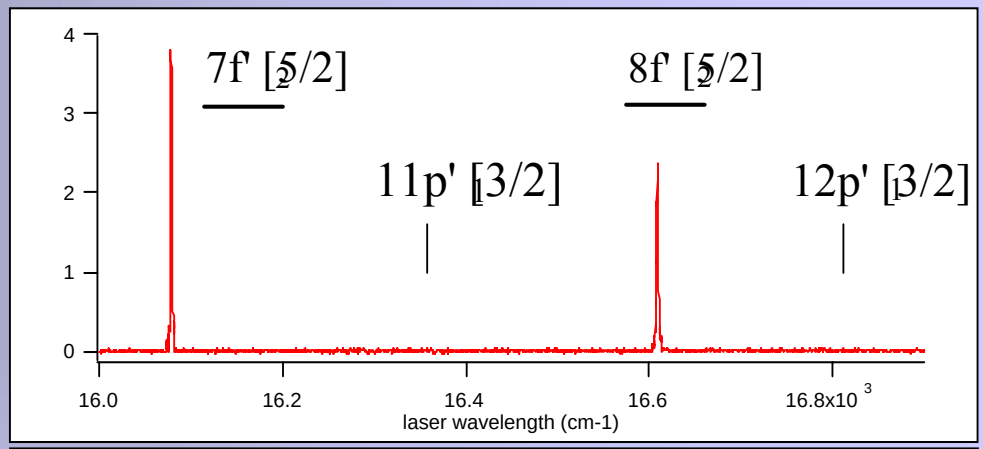
Xe* 5d --> 4f'

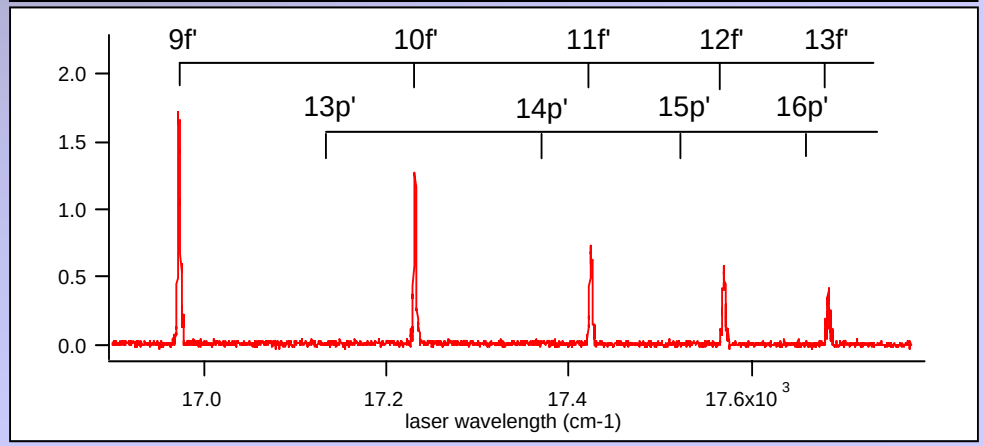


$\tau(5d) = 600ps$

$I_{pulsed} / I_{cw} = 75$

Xe* 6d --> nf', mp'

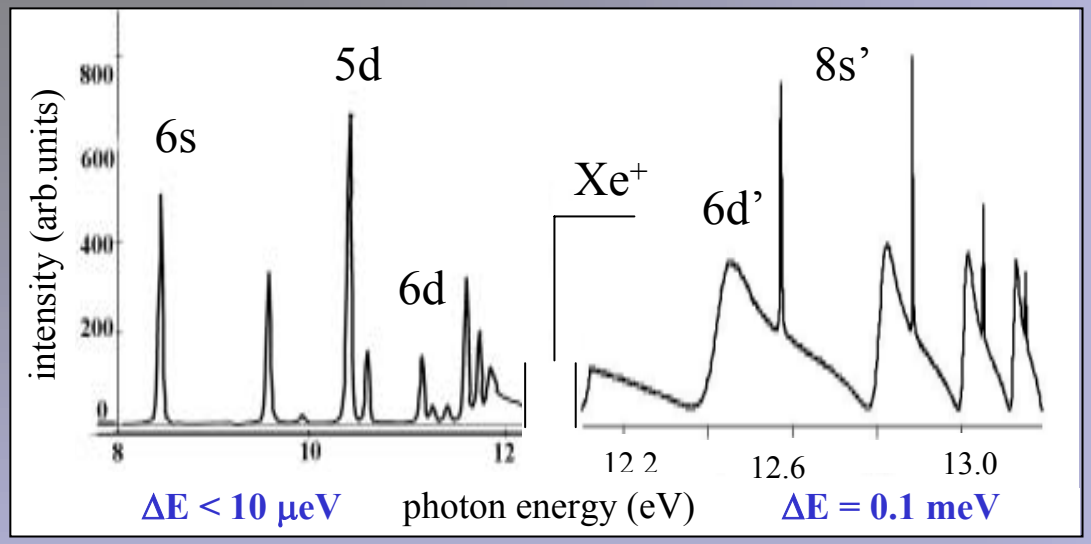


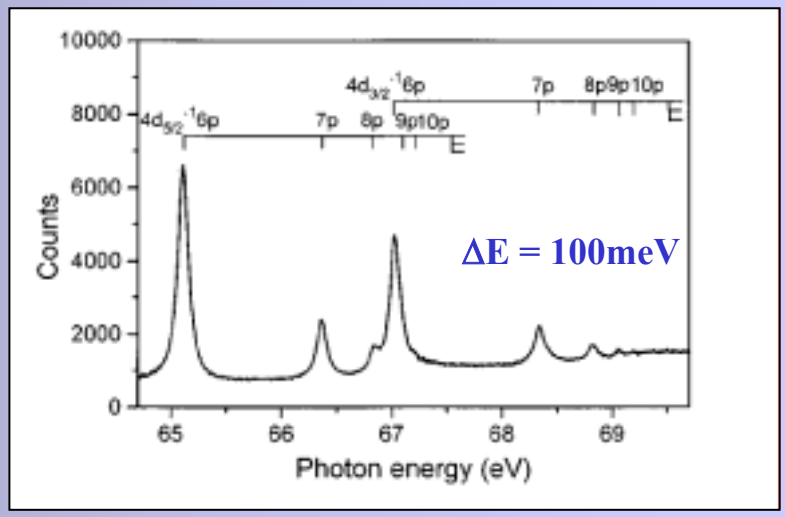


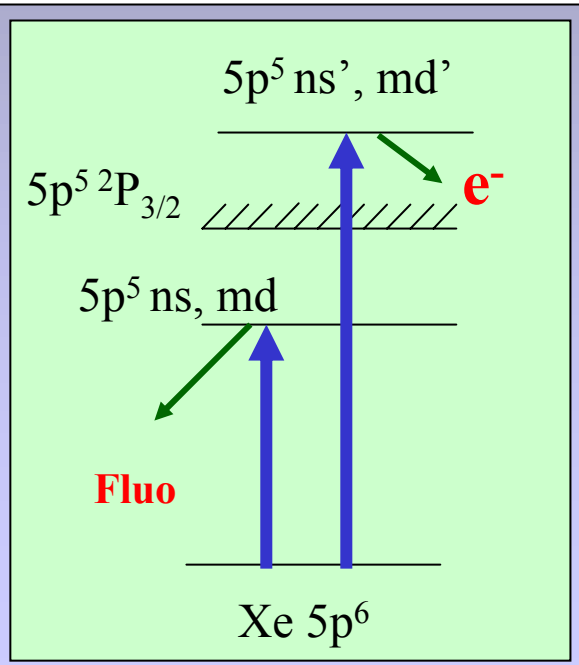
$\tau(6d) = 1.4$ ns

Photoexcitation in the valence shell

Core-excitation





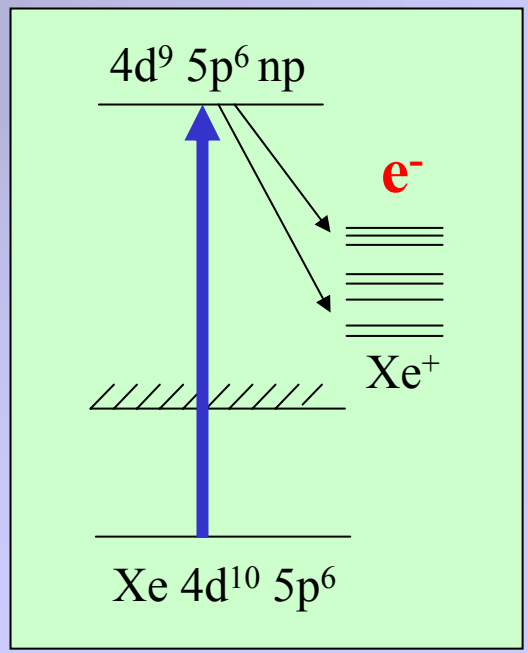


autoionisation

10ps

radiative relaxation

100ps - 10ns

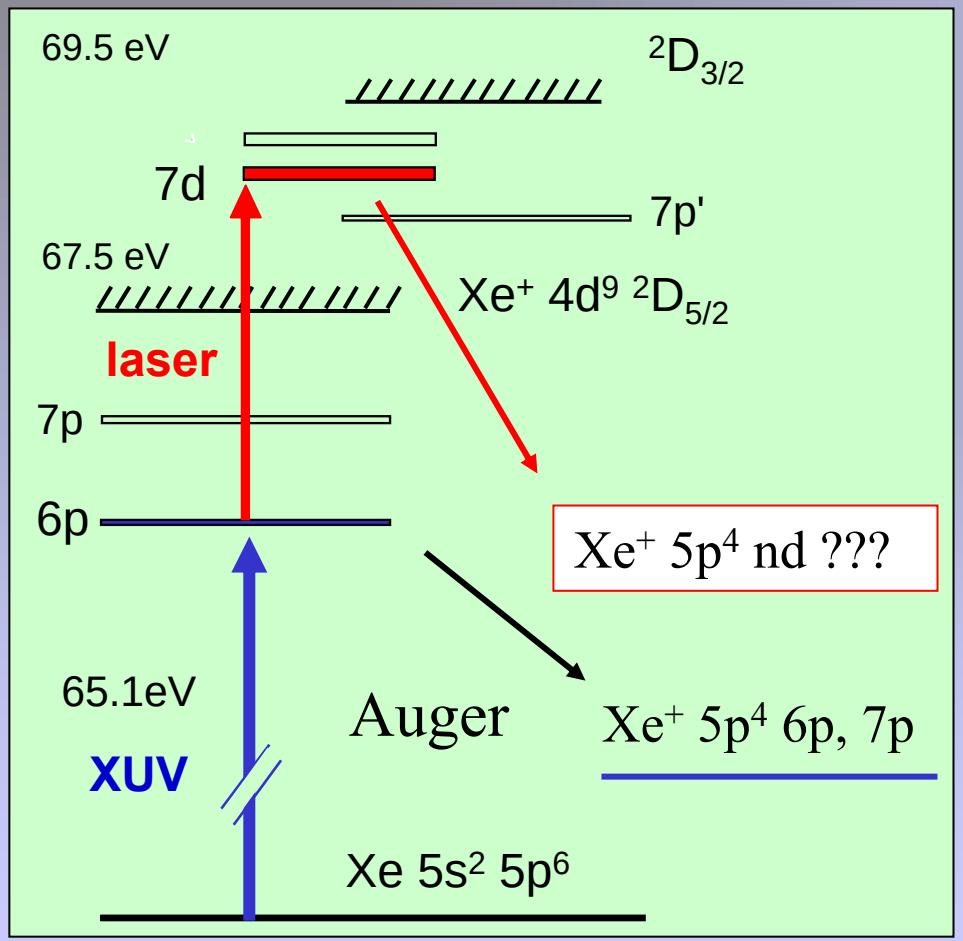


Auger decay

10fs

Two-photon (femtosecond) double-resonant excitation of autoionization states

Xe* $4d^9\ 5p^6\ 6p$ (Γ=0.11eV, τ = 6 fs) ----> Xe* $4d^9\ 5p^6\ 7d$

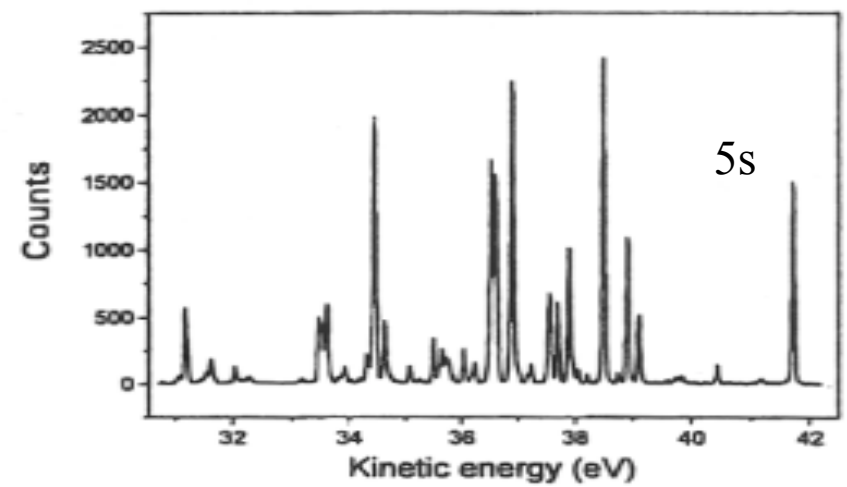


alternatives:

Li* 1s(2s2p ^3P) ^2P (2.6meV, 250fs)
----> Li* 1s($2p^2$) ^2D

Ar* $2p^5 4s$, 3d ($h\nu$ (exc.) = 245 eV)
----> Ar* $2p^5$ np, mf

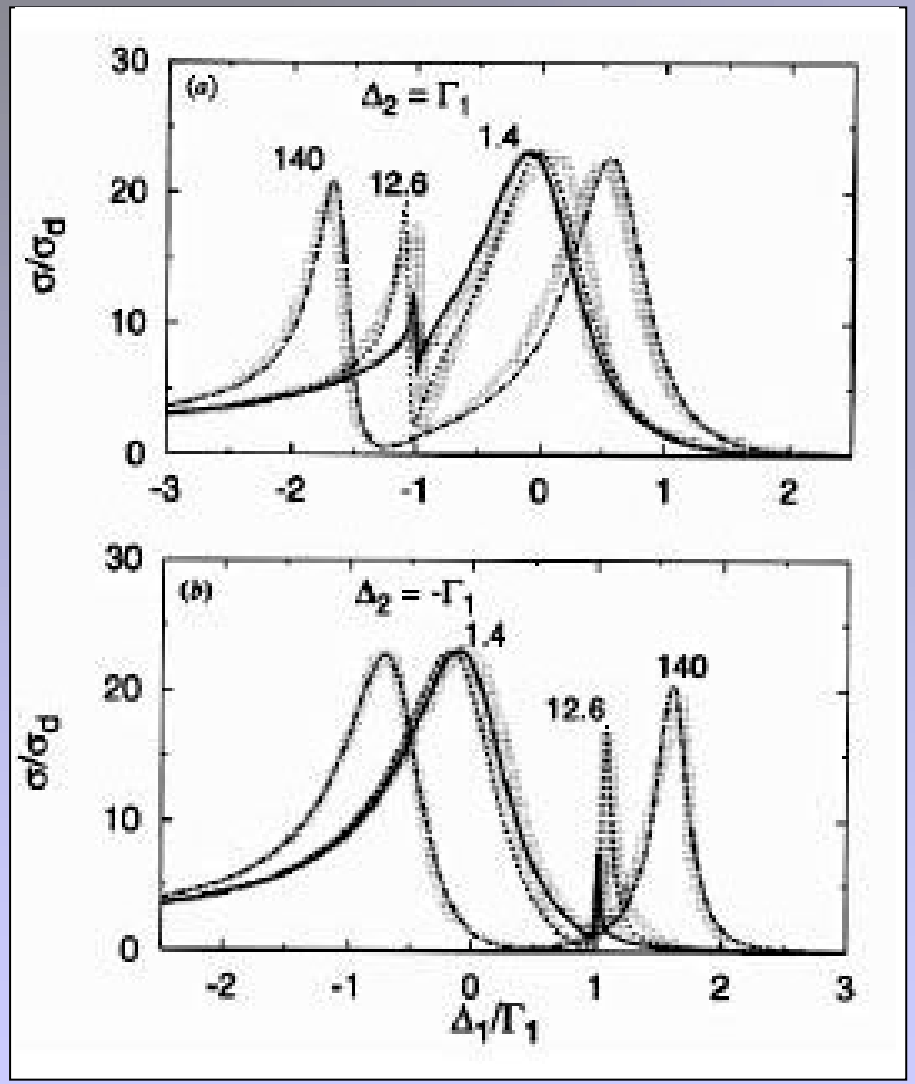
H. Aksela et al., Phys.Rev.A51,1291(1995)

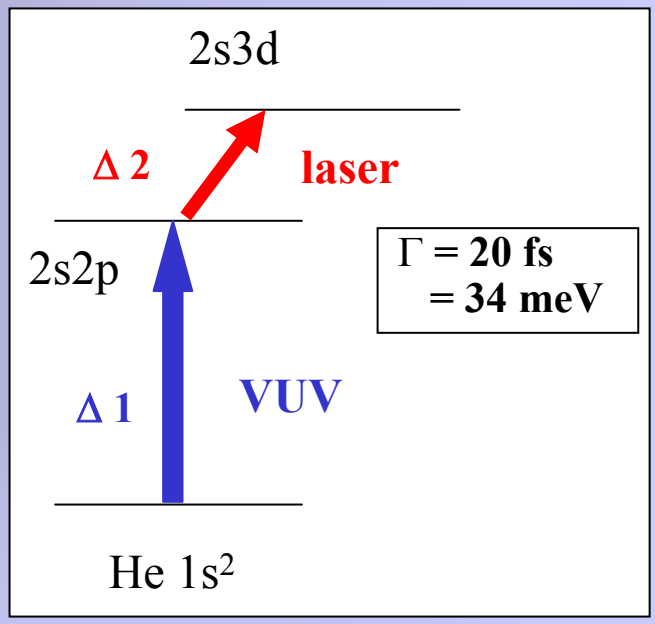


--> resonant Auger decay of even-parity AIS

Dynamics of autoionization states

coupling of two AIS by a (strong) laser field

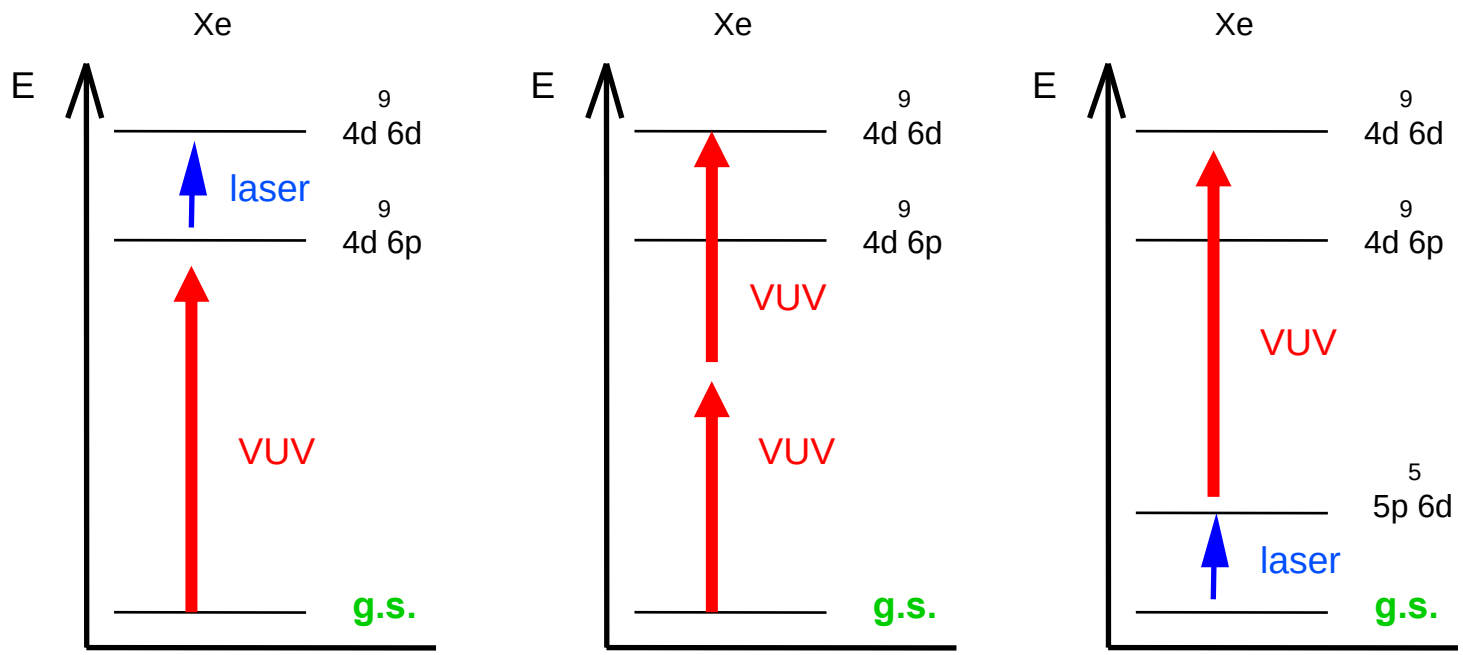




A.I. Magunov, I. Rotter, S.I. Strakhova, J. Phys. B32, 1489 (1999)
H. Bachau, Lambropoulos, Shakeshaft, PRA 34, 4785 (1986)

Dynamics of autoionization states

Effect of intermediate state and its lifetime



two-photon
double-resonant
VUV + laser

direct two-photon
VUV + VUV

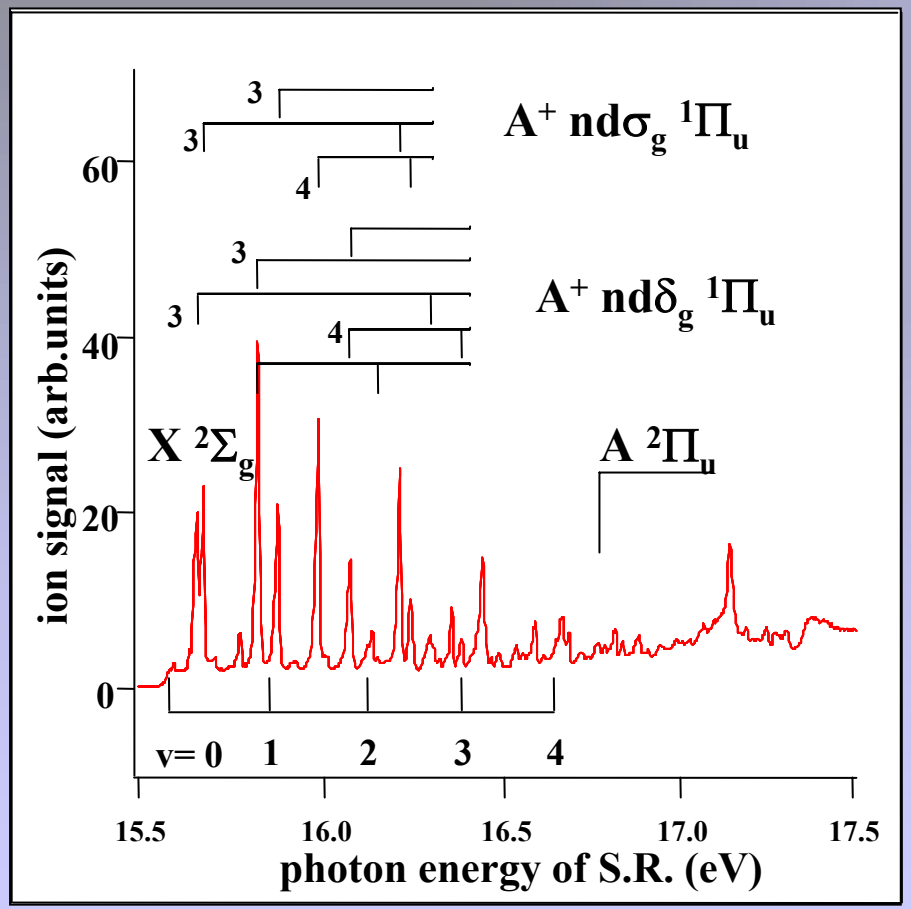
two-photon
double-resonant
laser (3hν) + VUV

Application:

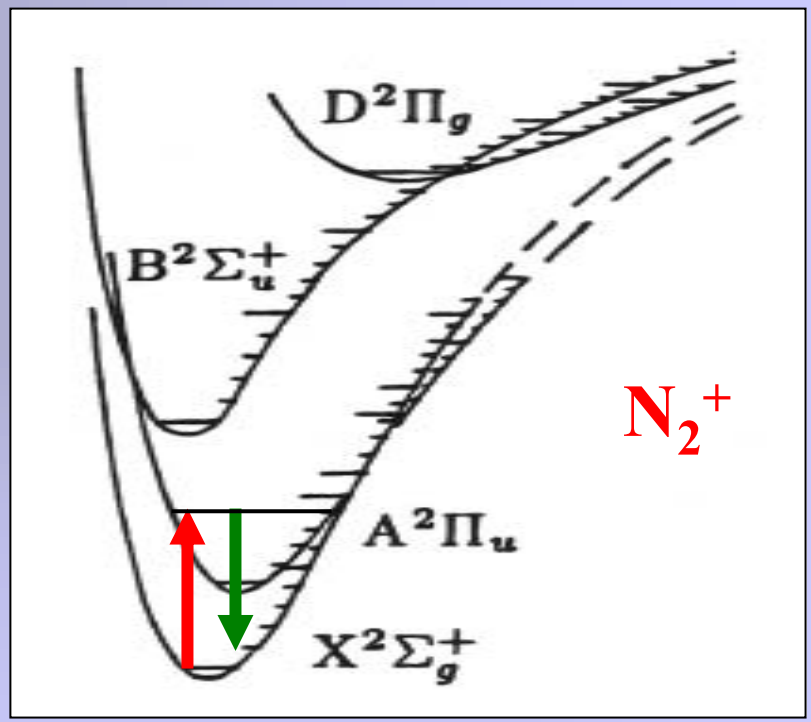
Dynamics of Molecular Fragmentation

Laser Induced Fluorescence spectroscopy on N_2^+ (SR + Laser)

N_2 + hν(S.R.) ---> N_2^* (Rydberg)
---> N_2^+ ($X\ ^2\Sigma$, v = 0, J'') + e^-



ion yield spectrum



excitation
v'' = 0 --> v' = 4
λ (laser) = 610nm

relaxation
v' = 4 --> v'' = 1
λ (fluo) = 710nm

Analysis of rotational structure of N_2^+ (SR + Laser)

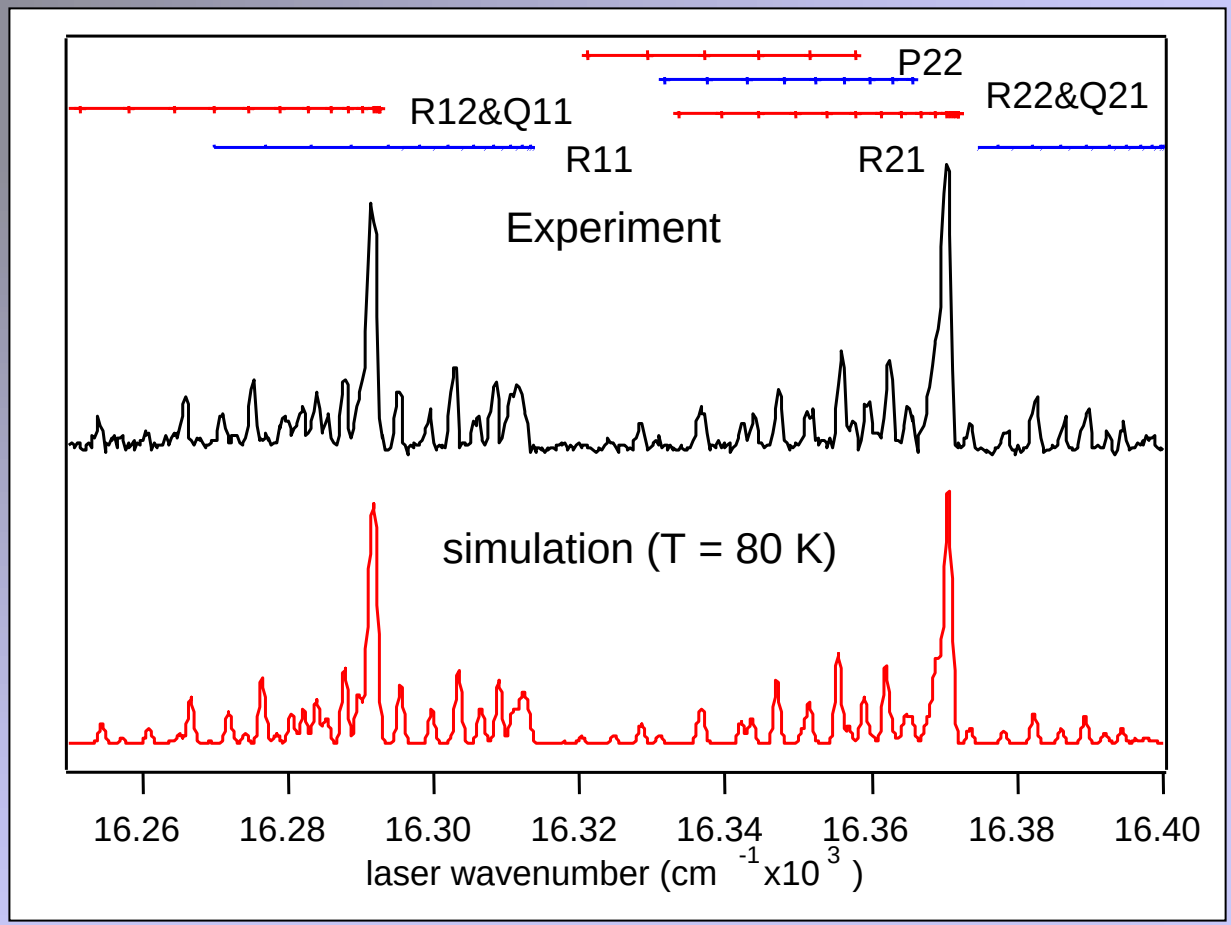
selection rules:

$\Delta J = -1, 0, +1$

P, Q, R branch

relative intensity:

exp (-B J(J+1) / kT)

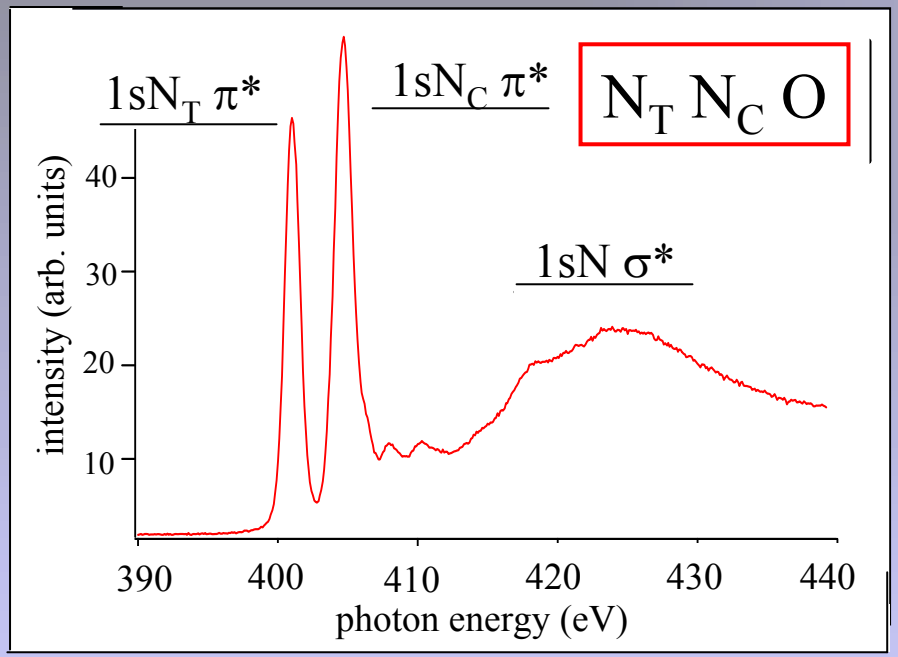


c.w. laser

high resolution laser spectroscopy on species prepared by synchrotron radiation

Resonant Core Excitations in molecules

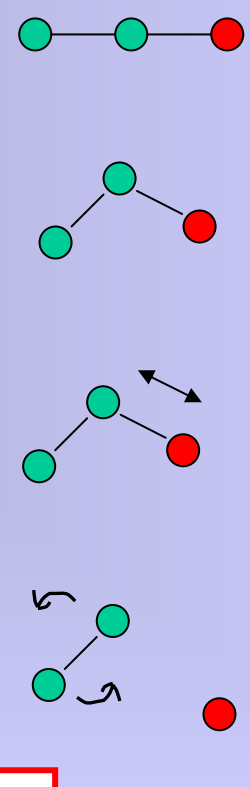
N_2O^{**} ---> N_2^+ + O



hν (401eV)

<10fs

laser



NNO

absorption

N*NO

Auger

NNO$^+$

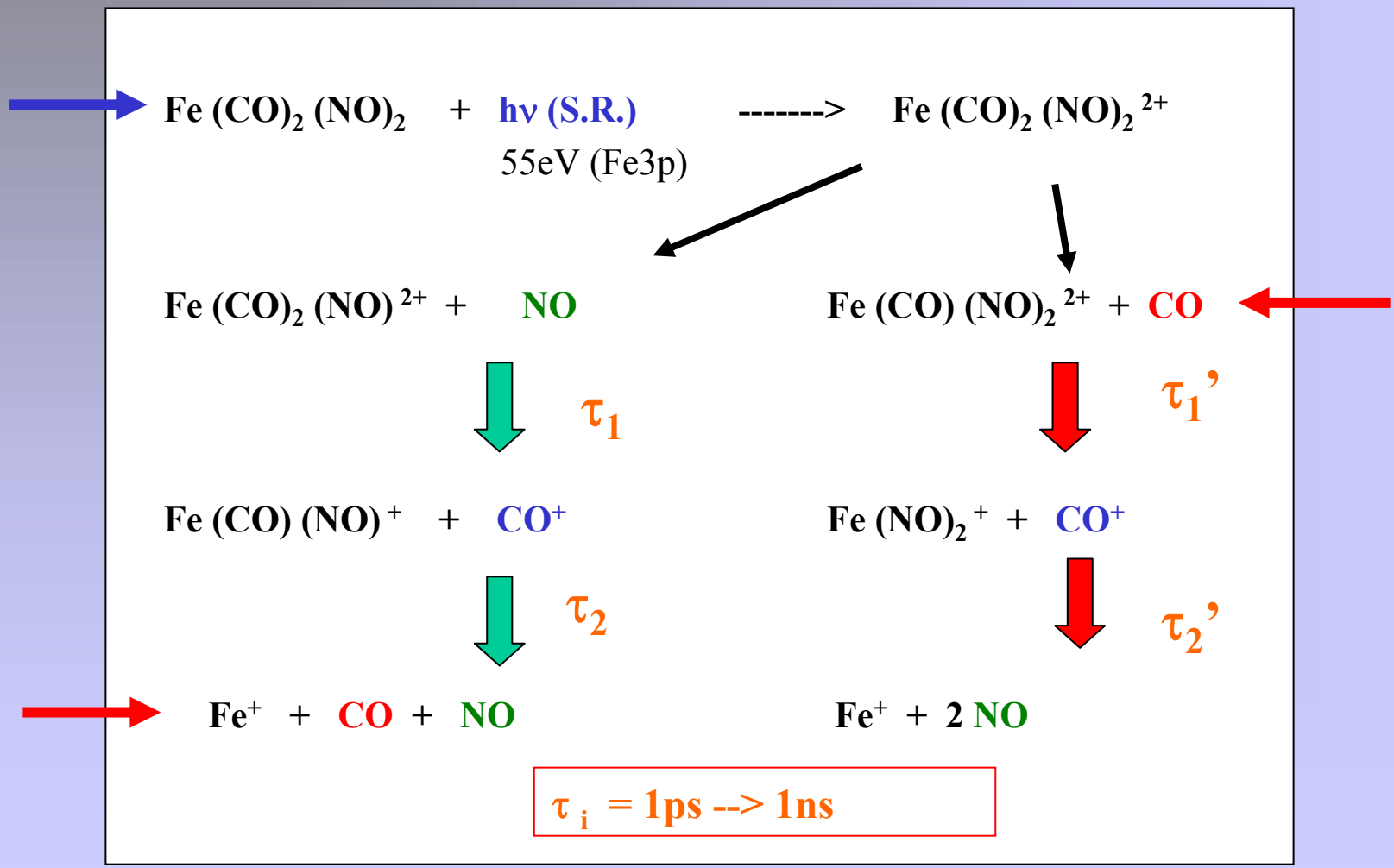
dissociation

NN$^+$ + O

coupling
electron <-> ion

Dissociation dynamics of core excited molecules

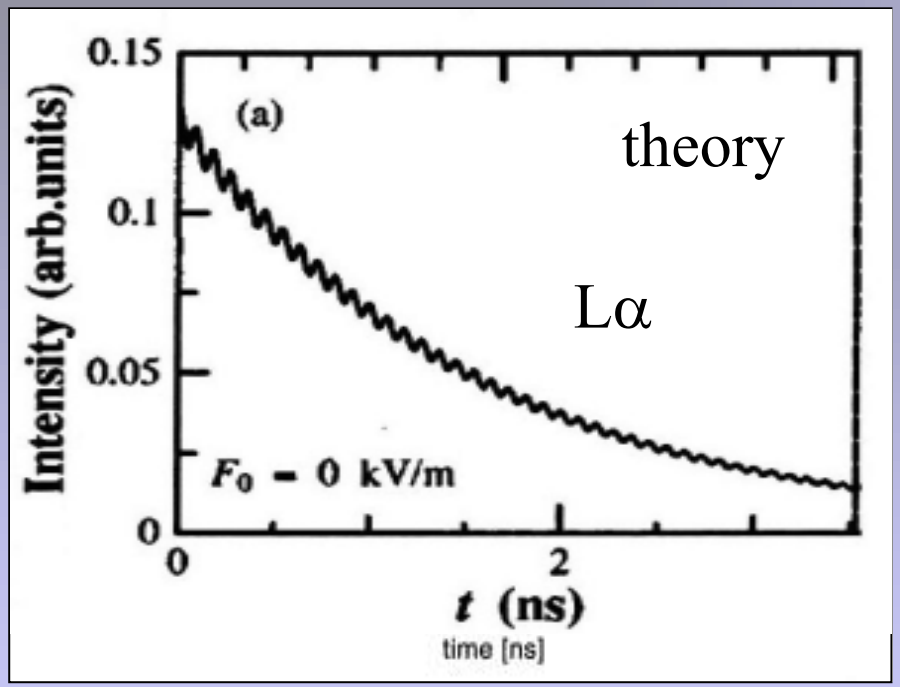
Time-resolved studies



Dynamics of molecular fragmentation

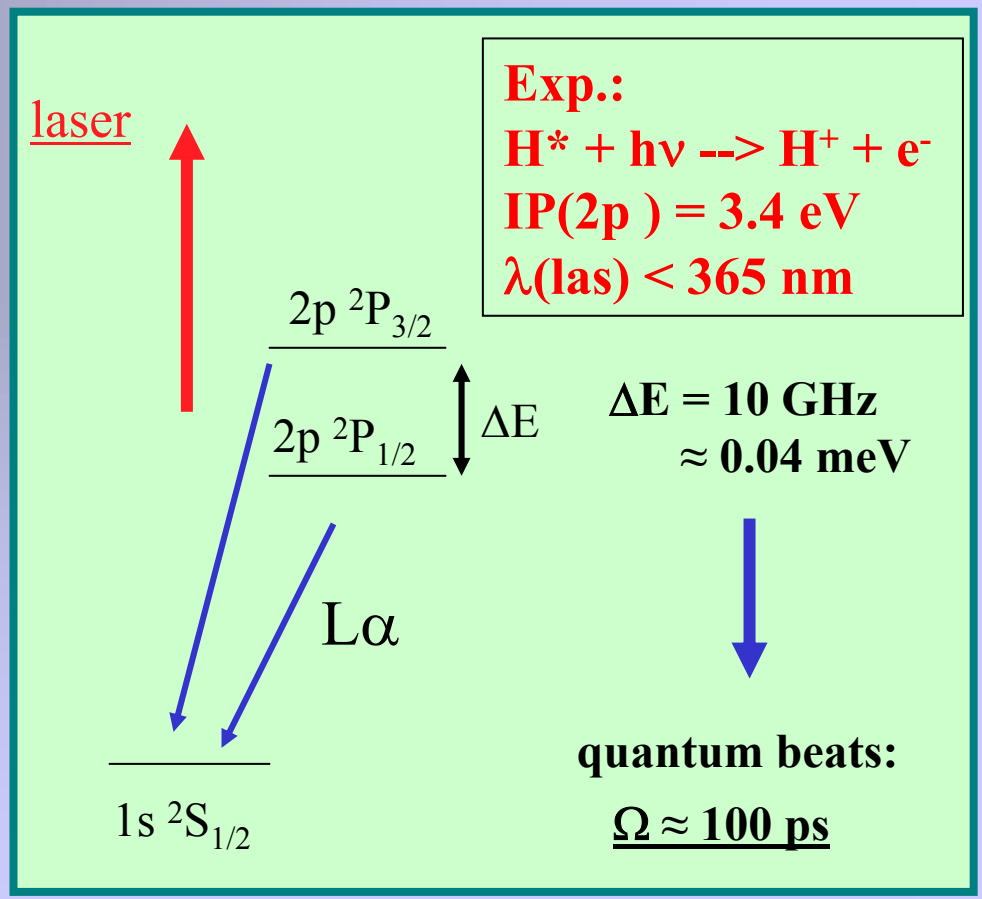
$H_2 + h\nu$ (FEL) ---> H + H* (2s , 2p, 3s, 3p …)

Fluorescence decay



Y. Kimura et al., Phys.Rev.A56, 4612 ('97), calculation, beamfoil-experiment

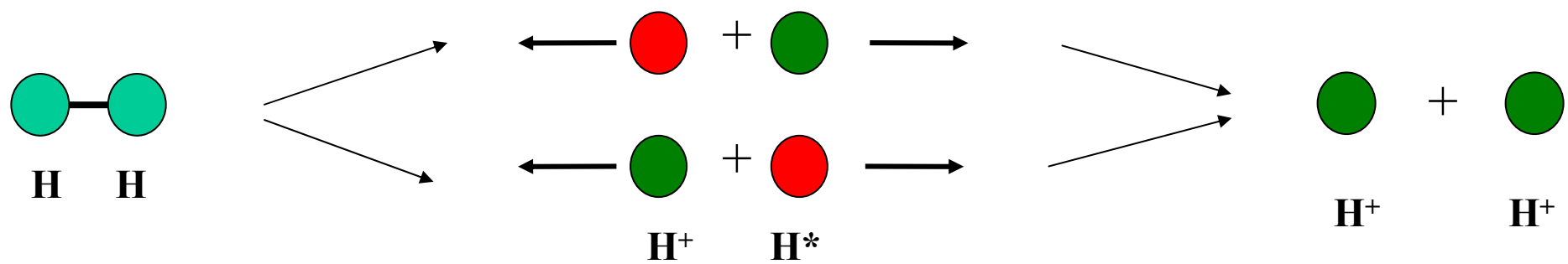
Ionization by laser



Interference effects in molecular fragmentation

Example: H_2

H_2 + **hν (FEL)** ---> H^+ + H* (2l , 3l …) ---> H^+ + H^+



Experiment:

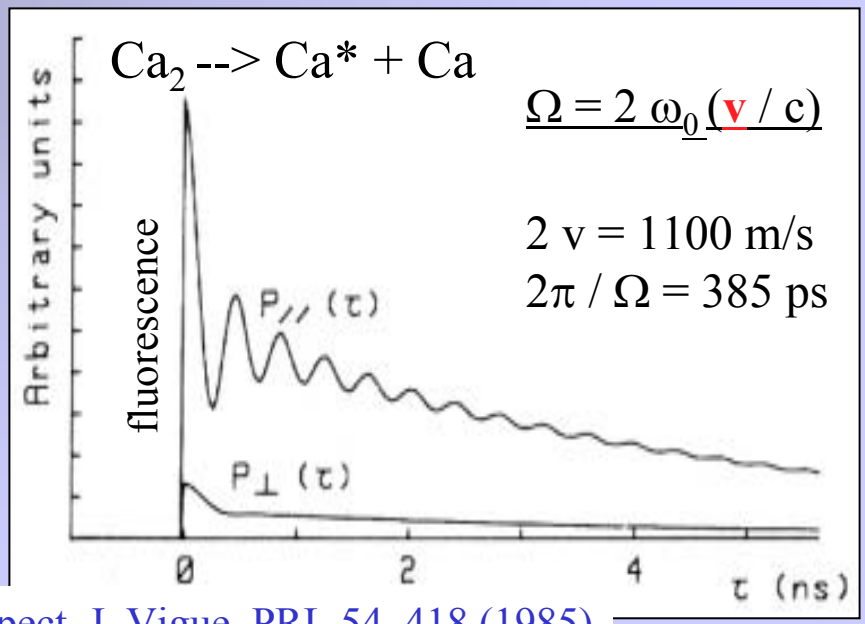
H*(3l) + hν (laser) --> H^+ + e^-

Doppler-Interference

λ(las) ≈ 800 nm --> E_{kin} (e^-) = 0.1 eV

hν(FEL) = 16.8 eV --> E_{kin} (H*) = 0.3 eV

$2\pi / \Omega = 2$ ps



P. Grangier, A. Aspect, J. Vigue, PRL 54, 418 (1985)

Conclusion

Pump-Probe experiments using femtosecond-X-Ray sources

Ultrafast dynamics of Photoionisation and Photodissociation

- **Characterization of FEL pulses**
  - **Cross correlation experiments**
- **Electronic Relaxation**
  - **Photoexcitation of Autoionization States**
  - **Coupling of Autoionization States**
  - **Dynamics of Rydberg Wavepackets**
- **Molecular Fragmentation**
  - **Time-Resolved Spectroscopy of Dissoziation**
  - **Interference Effects in Dissoziation**

Collaboration

SR + Laser Experiment

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Proposal: TESLA-FEL 2005

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