Coherent X-Ray-Optical Control of Nuclear Dynamics with Zeptosecond Phase-Stability

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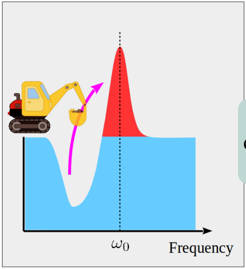
ISOQUANT SFB1225

Group of Jörg Evers at MPIK in Heidelberg

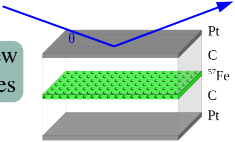


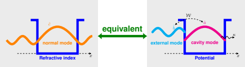
Coherent control of X-rays and nuclei

Nuclear quantum optics

X-rays excite new quantum regimes

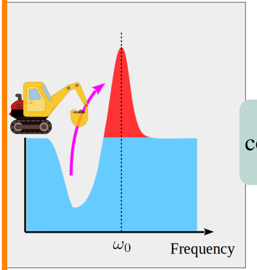
Understand fundamentals





Group of Jörg Evers at MPIK in Heidelberg

today



Coherent control of X-rays and nuclei

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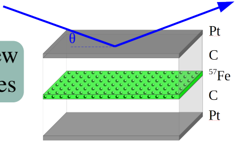
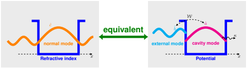
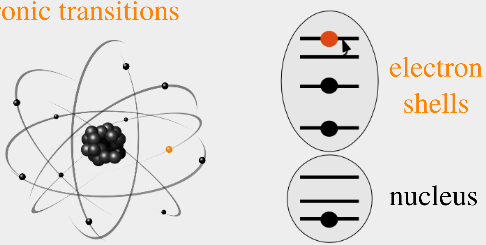


Understand fundamentals



From electronic towards nuclear quantum optics

Electronic transitions



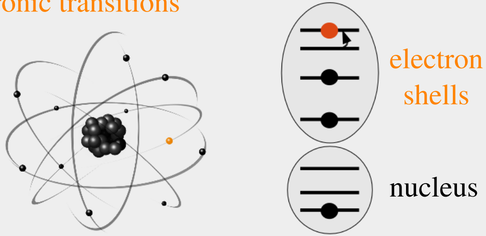
coherence, non-linearities, laser technology

↓

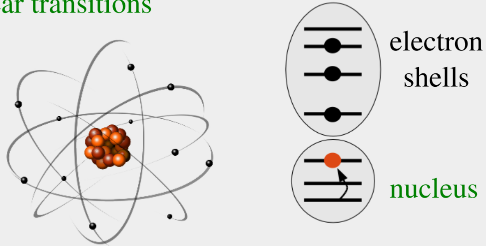
full quantum control

From electronic towards nuclear quantum optics

Electronic transitions



Nuclear transitions



coherence, non-linearities, laser technology

↓

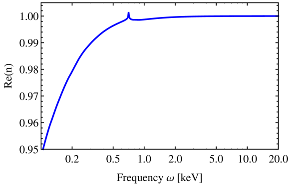
full quantum control

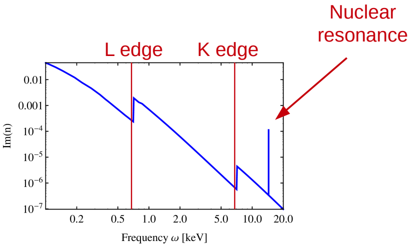
new approach to x-ray physics

new platform for quantum dynamics

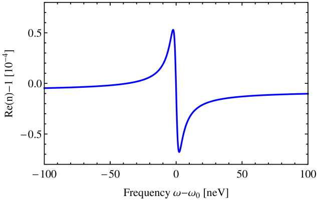
Mössbauer transitions

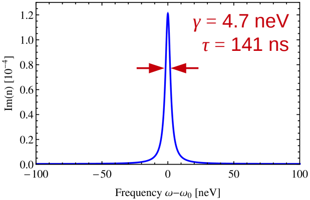
- Refractive index of ^{57}Fe





- Zoom in to ω_0 = 14.4 keV





Mössbauer transitions

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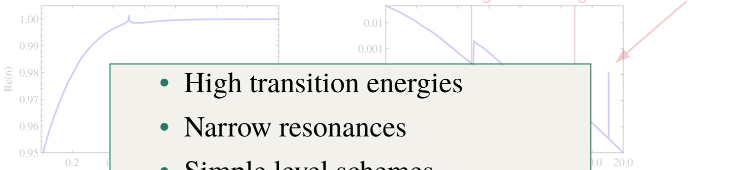
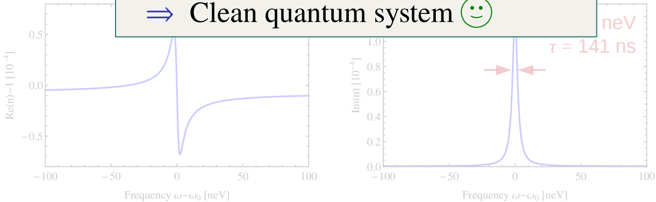


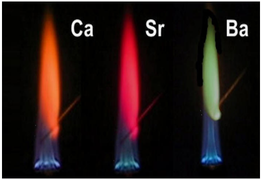
- Zoom in t



- High transition energies
- Narrow resonances
- Simple level schemes
- Essentially decoherence free

$\Longrightarrow$ Clean quantum system ☺

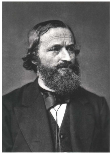
What can we learn from the visible range?



"Uncontrolled pump
&
passive observation"

1859

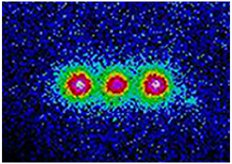
Bunsen



Kirchhoff

Coherence, Non-linearities,
Quantum effects, Lasers. . .

Today



"Full quantum control"

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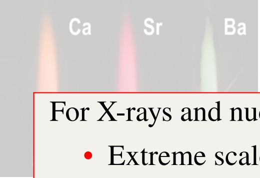

1859

For X-rays and nuclei:

- Extreme scales (energy, length, time)
- Source development
- Instrumentation limitations
- Relatively new field

$\Longrightarrow$ Nuclear/X-ray quantum optics is challenging!

Coherence, Non-linearities, Quantum effects, Lasers. . .

“Full quantum control”

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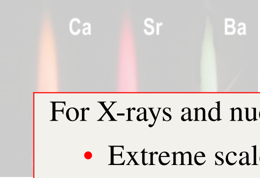

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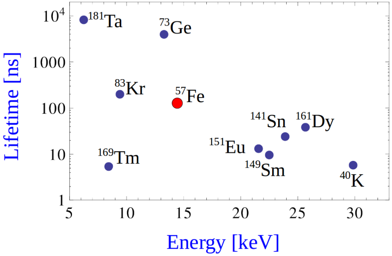
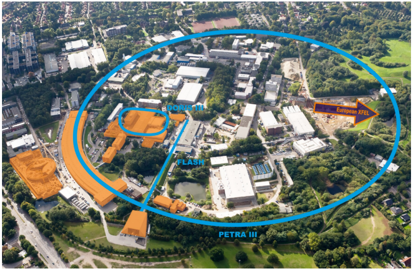
“Full quantum control”

Features and challenges

Mössbauer nuclei



X-ray sources



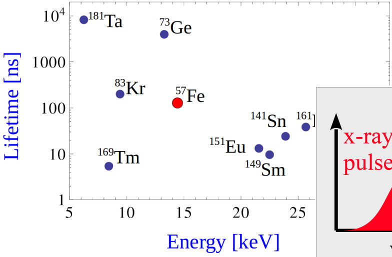
⇒ Narrow linewidths (μeV-peV)
⇒ Extreme quality factors

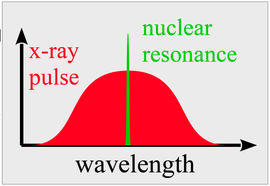
⇒ Broadband pulses
⇒ Low resonant intensity

⇒ **Linear regime of nuclear quantum optics**

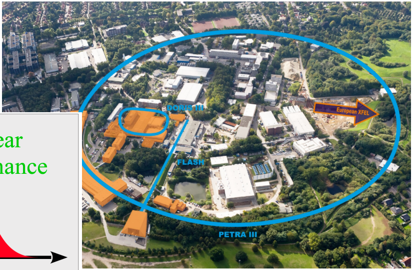
Features and challenges

Mössbauer nuclei





X-ray sources



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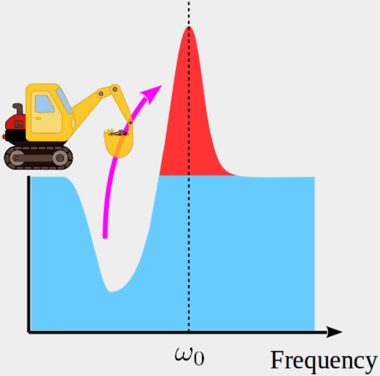
Overview

Project 1

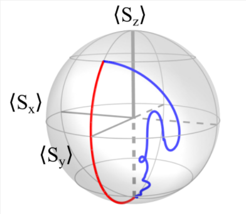
Spectral narrowing of X-rays



"Nuclei control X-rays"

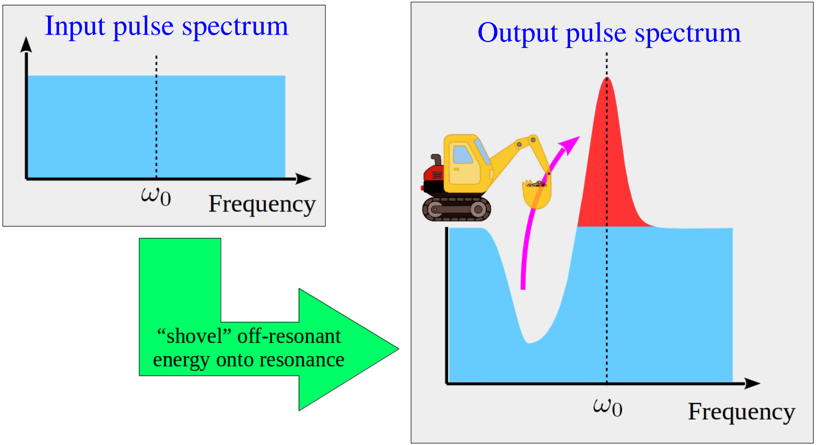
Project 2

Coherent control of nuclei



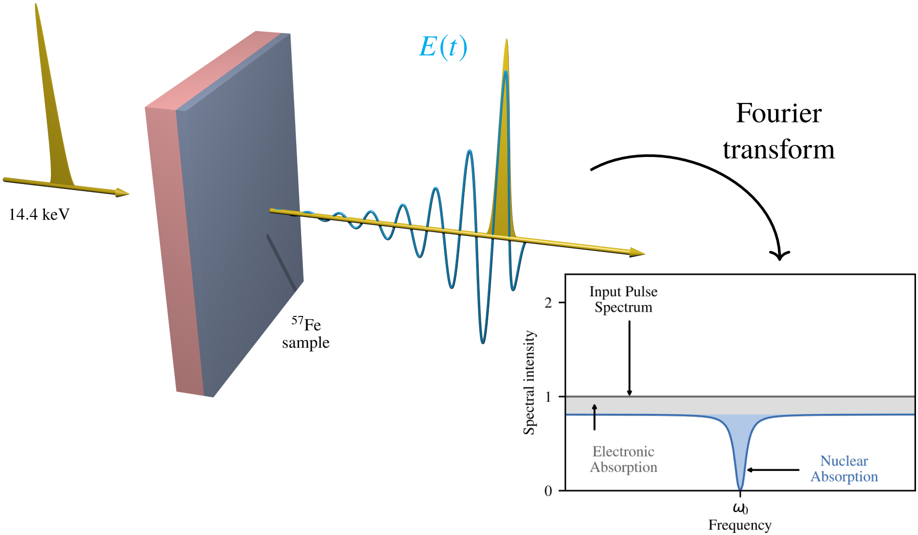
"X-rays control nuclear dynamics"

Result in a nutshell

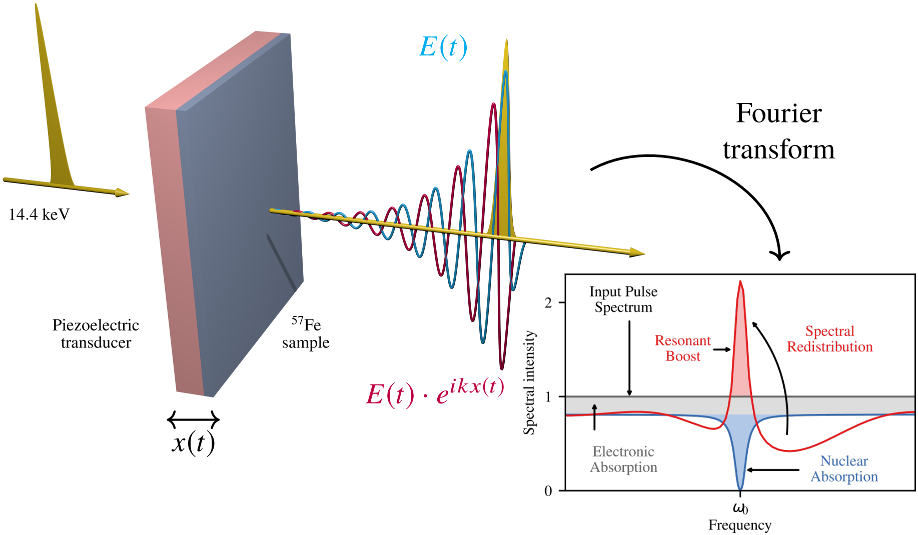


Heeg, Kaldun, Strohm, Reiser, Ott, Subramanian, DL, Haber, Wille, Goerttler, Rüffer, Keitel, Röhlsberger, Pfeifer, Evers, Science **357**, 375 (2017)

Setup: Nuclear forward scattering



Manipulation of the absorption line



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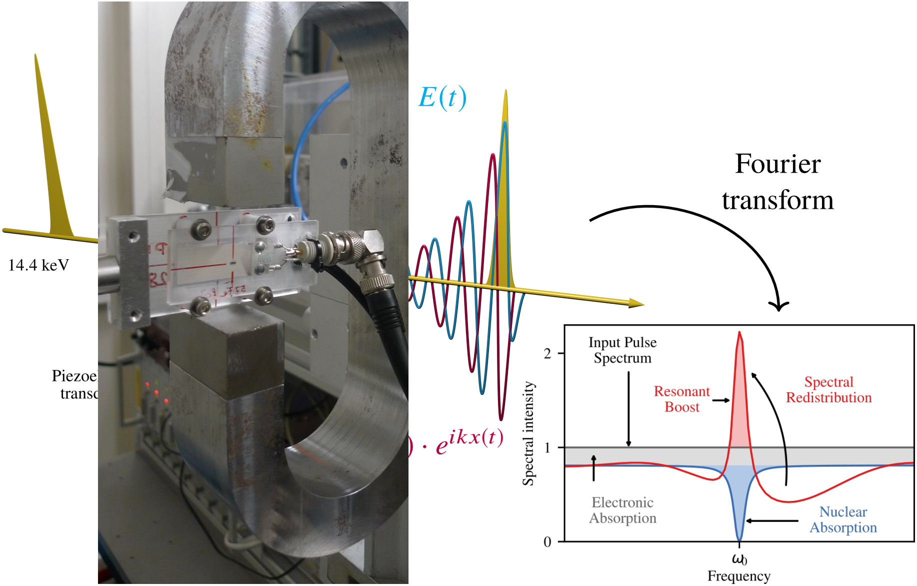

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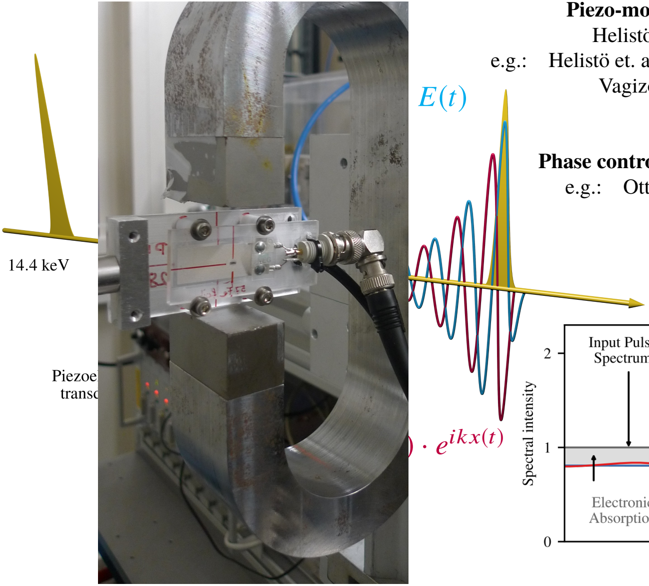


Piezo-motion well established concept:

Helistö, Kocharovskaya, Smirnov, . . .

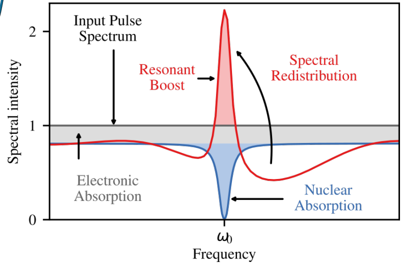
e.g.: Helistö et. al. *Phys. Rev. B* **34**, 3458 (1986)

Vagizov et. al. *Nature* **508**, 80 (2014)

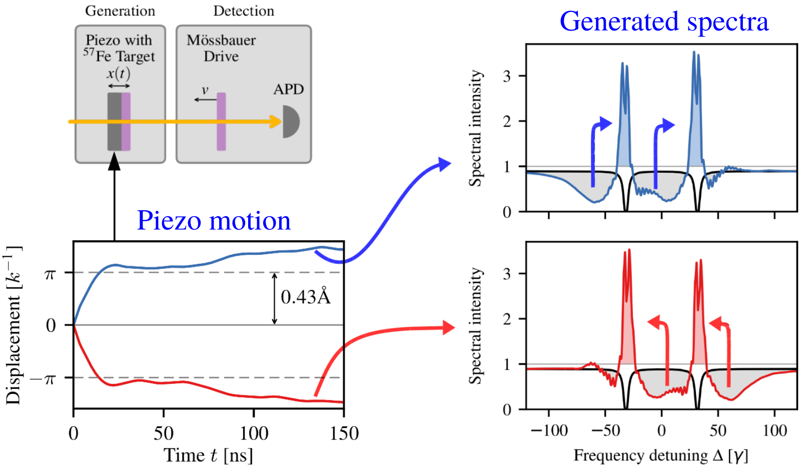
. . .

Phase control in attosecond spectroscopy:

e.g.: Ott et. al. *Science* **340**, 716 (2013)

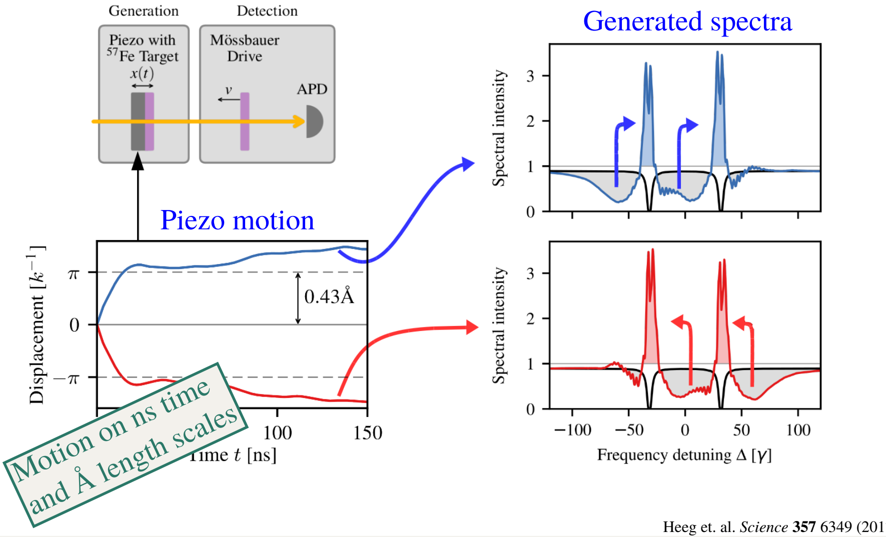


Experimental results



Heeg et. al. *Science* **357** 6349 (2017)

Experimental results

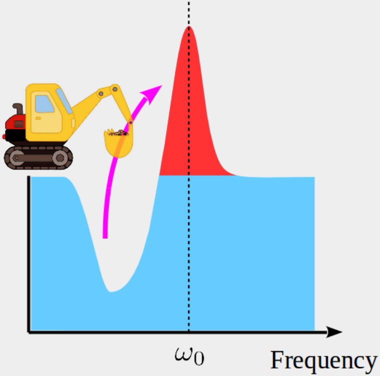


Heeg et. al. *Science* **357** 6349 (2017)

Overview: What's next?

Project 1

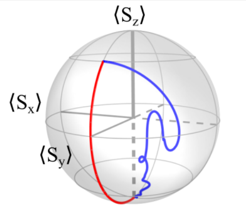
Spectral narrowing of X-rays



"Nuclei control X-rays"

Project 2

Coherent control of nuclei



"X-rays control nuclear dynamics"

Overview: What's next?

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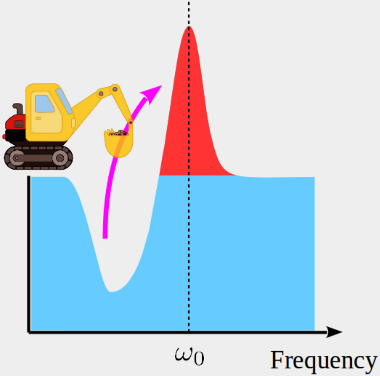
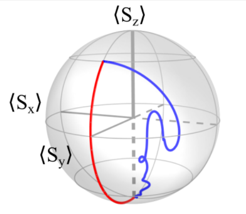
Spectral narrowing of X-rays



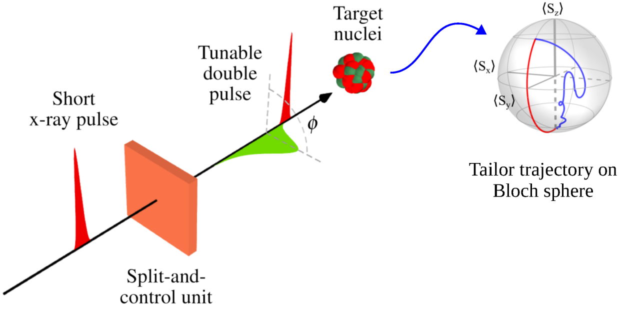
Project 2

Coherent control of nuclei



"Nuclei control X-rays" ⟶ "X-rays control nuclear dynamics"

What can be done with this new source?

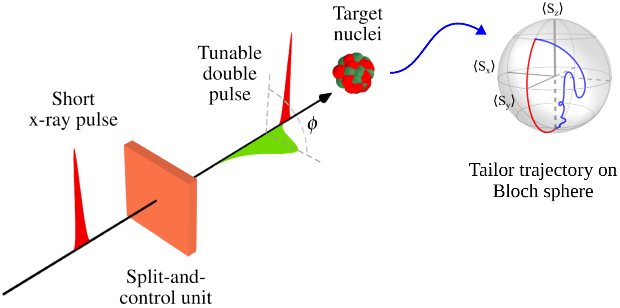


Split-and-control unit (SCU)

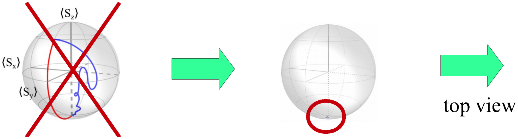
- Source for tunable x-ray double-pulses
- Control phase, frequency, . . . of second pulse relative to first one

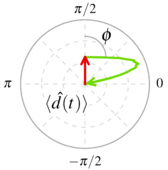
$\Rightarrow$ Coherent control of nuclear excitation

What can be done with this new source?



Here: focus on low excitation at synchrotrons





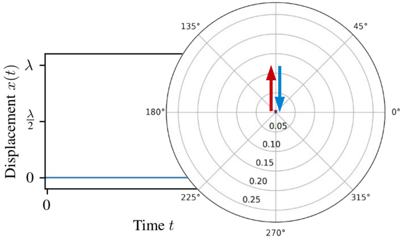
Two special cases of control

- “Stimulated emission of excitons”

  Two pulses with opposite phase:

  First preparation pulse excites exciton

  Second control pulse de-excites target

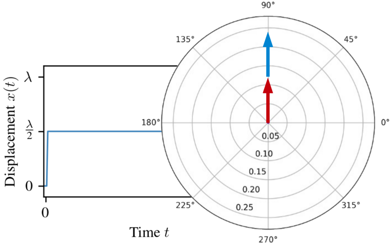


- “Coherent boost of excitation”

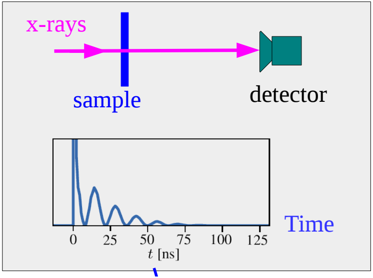
  Two pulses with same phase:

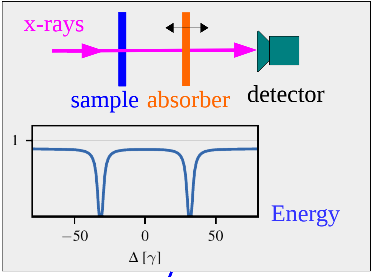
  First preparation pulse excites exciton

  Second control pulse further excites target



How to observe?





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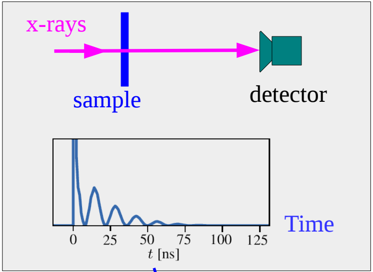
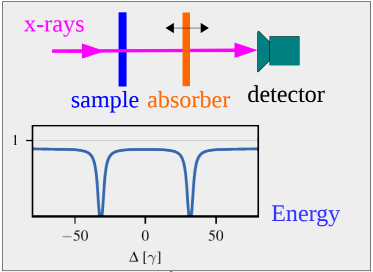
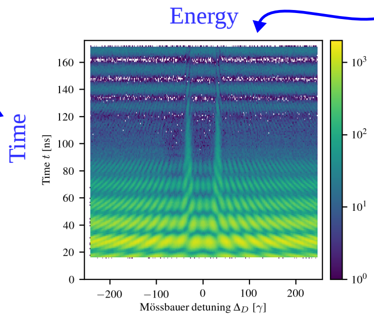




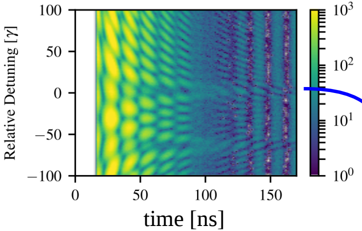
correlate properties



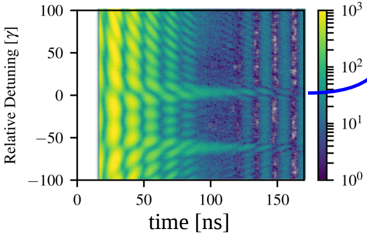
tomographic information encoded in interference pattern

Experimental results (at ID 18, ESRF)

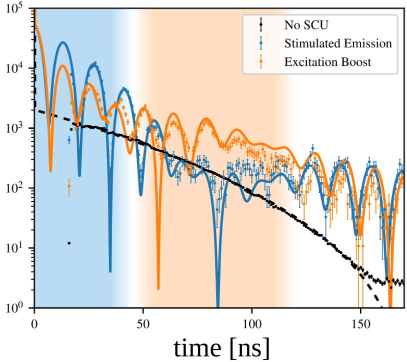
“Stimulated emission of excitons”



“Coherent excitation boost”



slices at detuning = 0



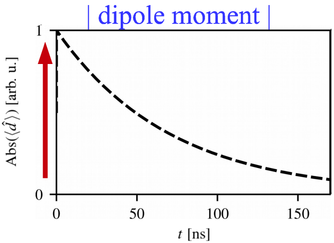
“Rule of thumb”:

SE → More intense at early times

Boost → More intense at later times

K. P. Heeg et al. *submitted*

Nuclear dipole moment



Regular decay without second pulse

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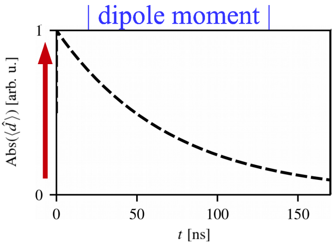
Nuclear dipole moment

Excitation boost
Stimulated emission



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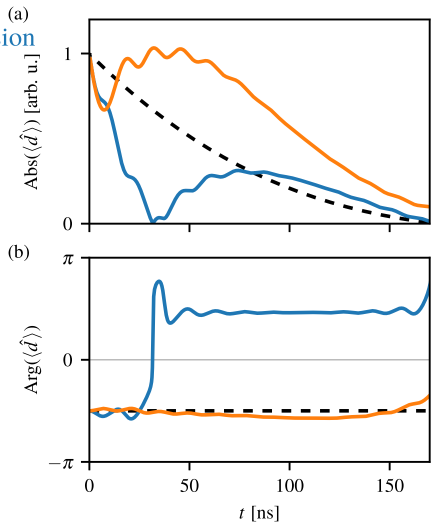

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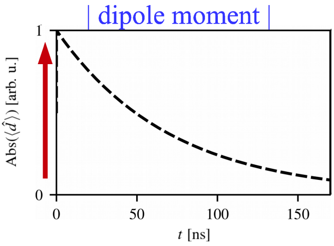

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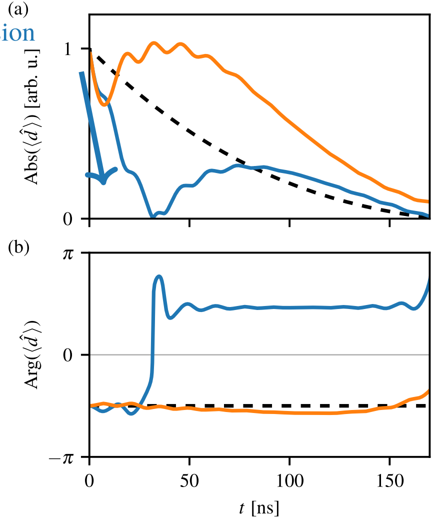

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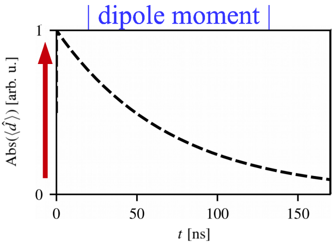

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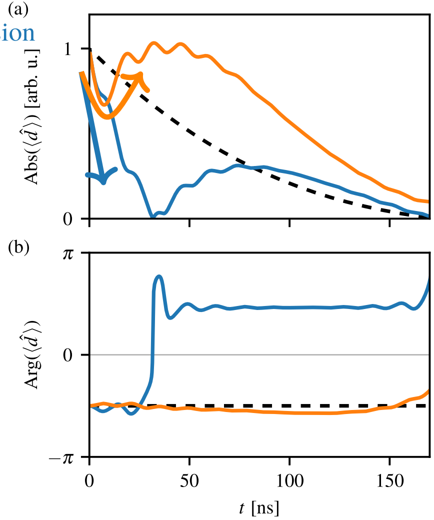

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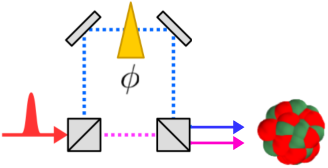
Why is the control so stable?

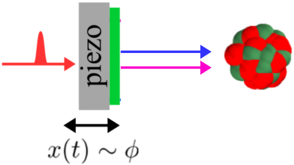
"Conventional approaches"

- Interfering pathways spatially separated
- Geometry must be stabilized throughout the entire long accumulation of statistics

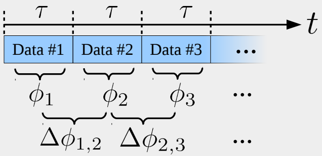


Piezo control with mechanical motion

- Interfering pathways coincide in space
- Control depends on motion relative to the geometry at the time of excitation
- Geometry only needs to be stable for a ~ 200 ns measurement interval after each x-ray pulse
- All other drifts / noise do not matter!



Phase stability



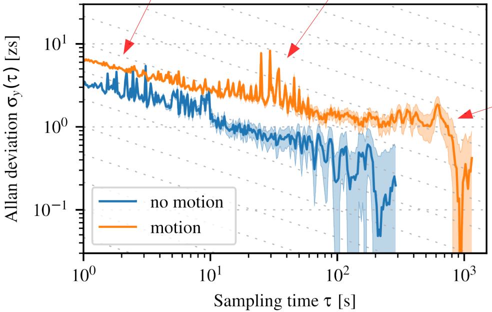
RMS → $\sigma_y(\tau)$

RMS instability of two measurements τ apart

Allan *Proc. IEEE* **2**, 221 (1966)

$\sim 1/\sqrt{\tau}$

artefact due to dead times in data acqusition electronics



Systematics due to motion?

Excitation boost

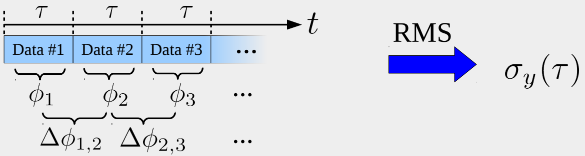
Stimulated emission

X-ray period 287 zs

Measurement time ~ 45 min

few-zeptosecond phase stability!

Phase stability



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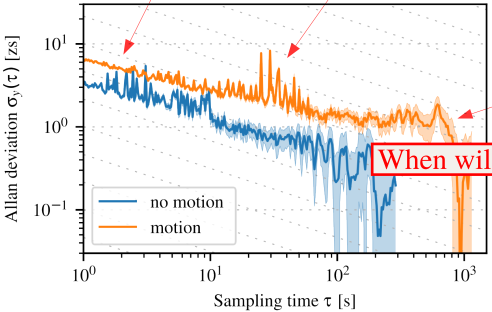


Systematics due to motion?

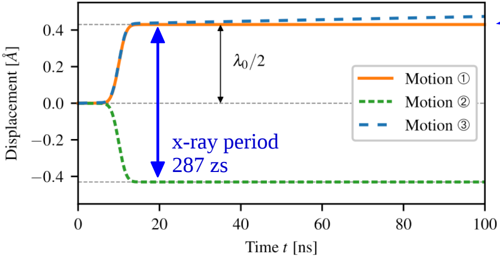
When will it saturate?

X-ray period 287 zs

Measurement time ~ 45 min

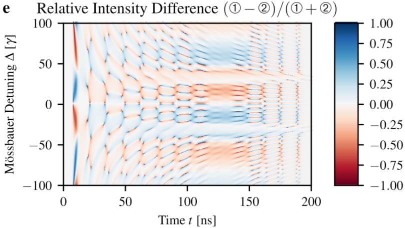
few-zeptosecond phase stability!

Tomography of the piezo motion

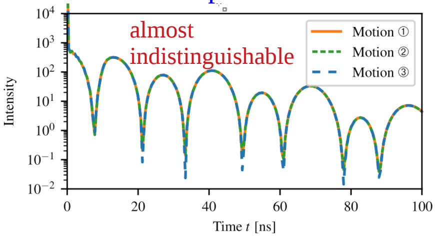


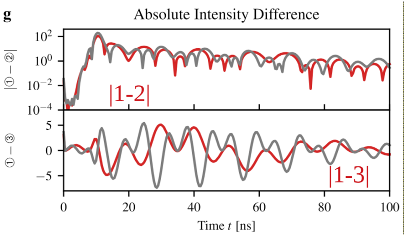
Simulated phase error ~ 25 zs

2D spectra



1D spectra





K. P. Heeg et al. *submitted*

Outlook

More resonant photons

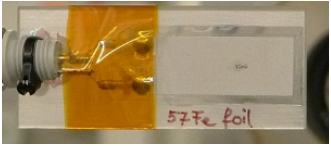
→ faster measurements
→ smaller samples

Adaptive X-ray optics

X-ray quantum optics

→ control of dynamics
→ simulate control fields

much more. . .



Track motion on sub-Angstrom scales

Ideas and concepts are valid beyond x-rays

→ e.g. attosecond spectroscopy by T. Pfeifer et al @ MPIK

Multi-dimensional X-ray spectroscopy

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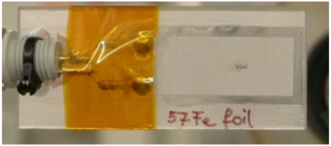

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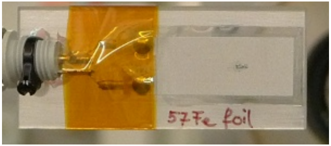

Track motion on sub-Angstrom scales

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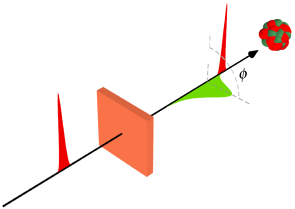
Multi-dimensional X-ray spectroscopy

X-ray phase detection?

Summary & Acknowledgements

- Control of light-matter interaction via mechanical motion
- Enhance resonant intensity of given x-ray pulses via spectral redistribution
- Generation of tunable phase-coherent x-ray double pulses
- Control of nuclear dynamics between “stimulated emission” and “excitation boost” with zepto-second stability



The team:

Heeg, Kaldun, Strohm, Reiser, Ott, Subramanian, Lentrodt, Haber, Wille, Goerttler, Rüffer, Keitel, Röhlsberger, Pfeifer, Evers, Science **357**, 375 (2017) + submitted

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