Is strong excitation feasible in ensembles of Mössbauer nuclei?

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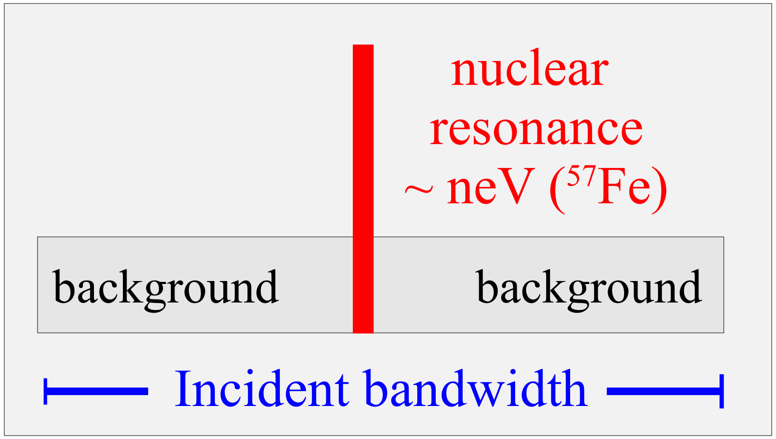
XFELO retreat, SLAC, 30 June 2016

How can XFELO make a difference?

Problem *and* feature: Narrow resonance



nuclear resonance

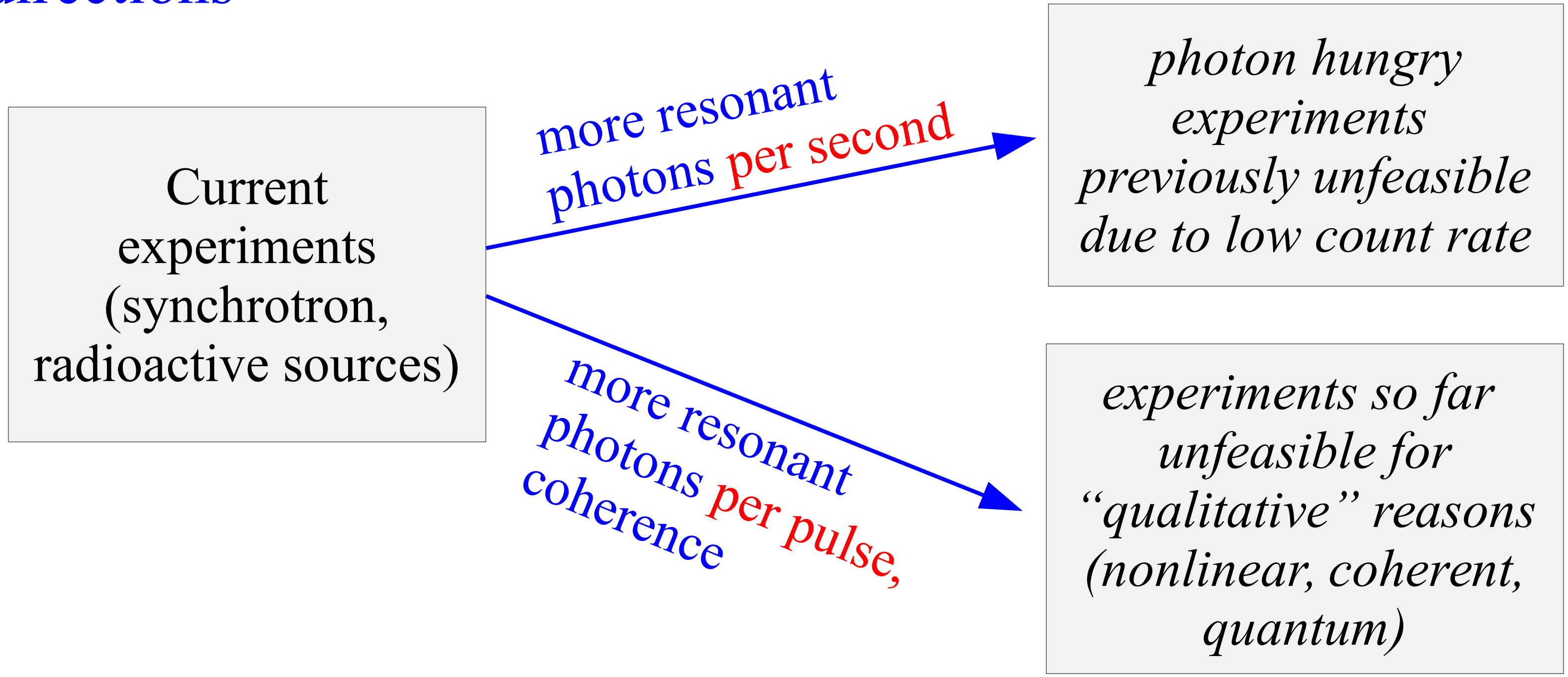
~ neV (^{57}Fe)

background

background

Incident bandwidth

Two directions



Current experiments (synchrotron, radioactive sources)

more resonant photons per second

photon hungry experiments previously unfeasible due to low count rate

more resonant photons per pulse, coherence

experiments so far unfeasible for “qualitative” reasons (nonlinear, coherent, quantum)

Qualitatively new regimes

Beyond single excitation :

- Quantum effects (2-photon entanglement, correlations)
- “Correlation spectroscopy” (c.f. talk by R. Santra) → $G^{(2)}$, dynamics (maybe also of “host material”)
- Nonlinear light-matter interaction (e.g. coherent enhancement of nonlinear index of refraction)

Strong excitation / full inversion :

- Strong control fields for advanced quantum optical schemes
- Excited state dynamics, out-of-equilibrium aspects
- Nonlinear spectroscopy
- Macroscopic population transfer (nuclear structure, batteries, sample preparation)

What are “qualitatively different” conditions?

Benchmark proposed here:

Single excitation
per shot

Goal

Full inversion
of ensemble

Why is this useful? Because population inversion...

- requires large number of resonant photons per pulse
- requires temporal coherence
- may benefit from phase coherence of subsequent pulses

Key features
of
XFELO

What is the current state of the art?

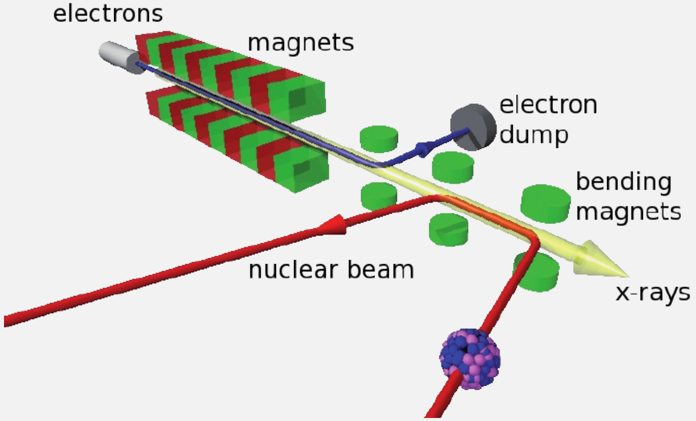
- Fairly long history [see e.g. review S. Matinyan, Phys. Rep. 298 199 (1998)], also related to gamma-ray laser

- Different excitation schemes were considered
  - nuclear beams
  - nuclei in solid state targets
  - XFEL / XFELO
  - high-power IR/optical lasers
  - …

- So far only low excitation predicted even for favorable x-ray parameters

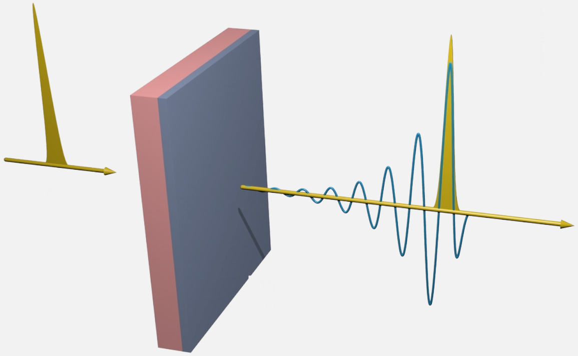
- Another problem: How to reliably detect inversion with messy / unstable x-ray pulses?

XFEL and nuclear beams



T. Bürvenich, J. Evers, C. H. Keitel, PRL 96, 142501 (2006)

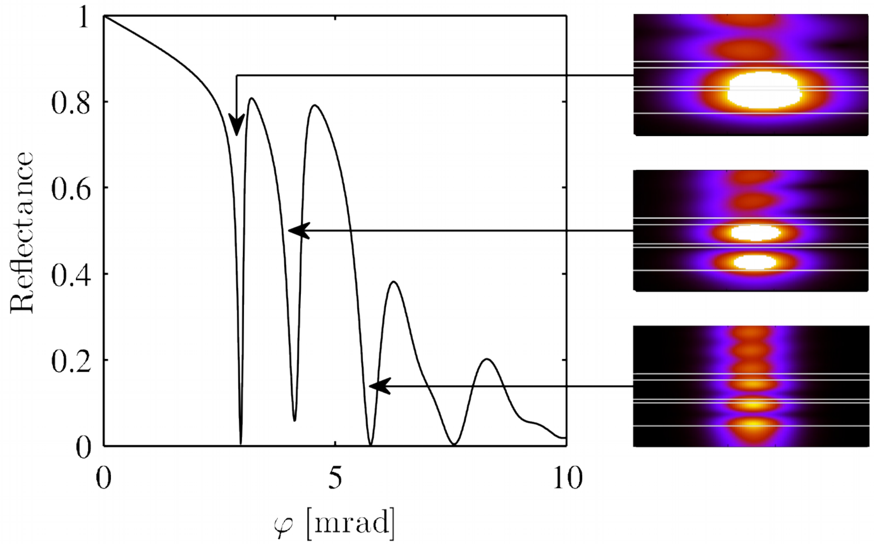
XFEL/XFELO and solid state targets

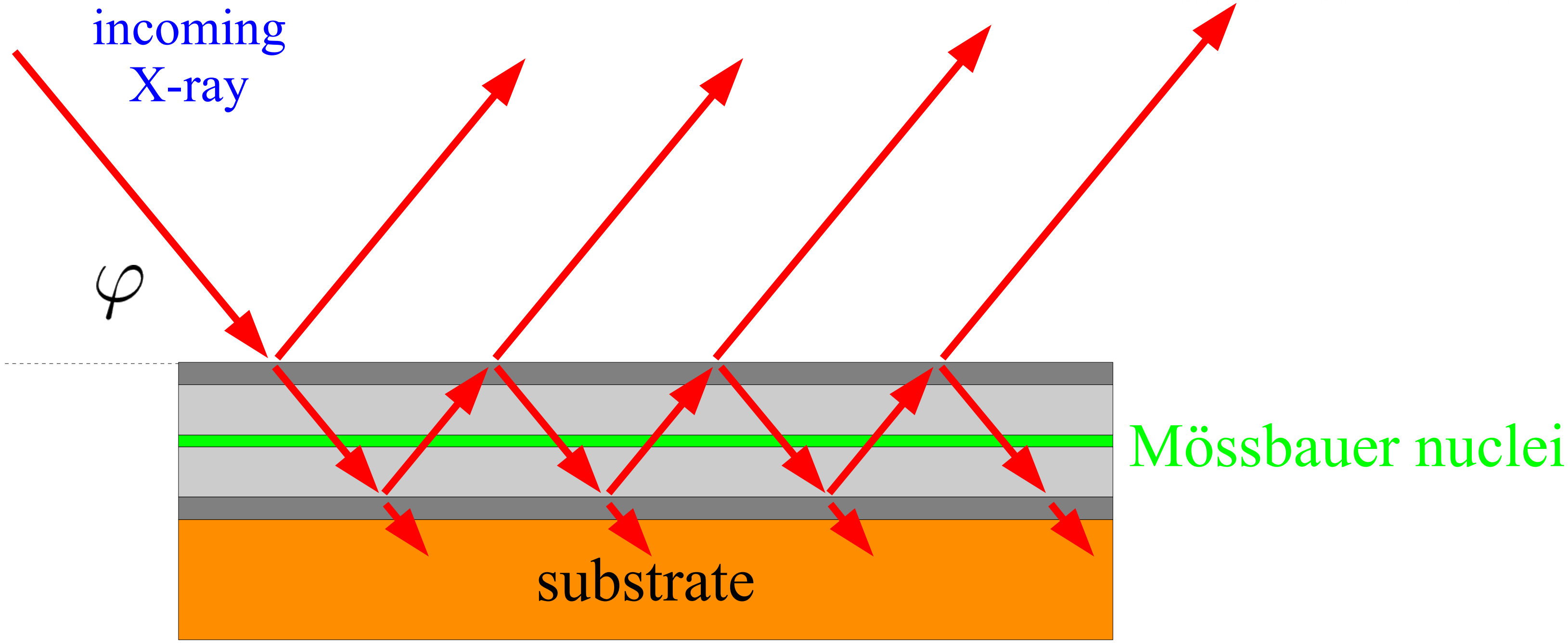


A. Junker, A. Palffy, C. H. Keitel, New J. Phys. 14, 085025 (2012)

What is different in our approach?

- Starting point: x-ray waveguides
- Promising, since cavity enhancement of interaction together with coherence-based effects have been demonstrated *)
- Systematically optimize the nuclear target





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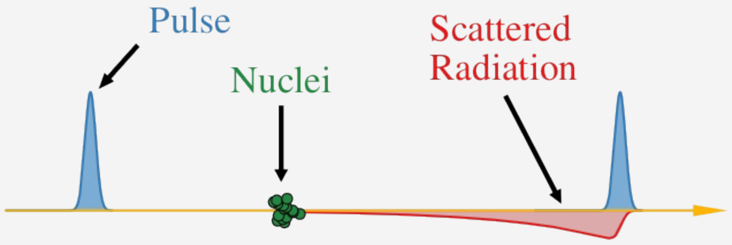
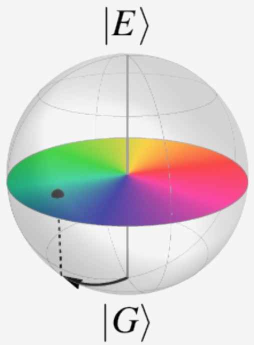
*) Heeg, Haber, Schumacher, Bocklage, Wille, Schulze, Loetzsch, Uschmann, Paulus, Rüffer, Röhlsberger, Evers, Phys. Rev. Lett. 114, 203601 (2015)

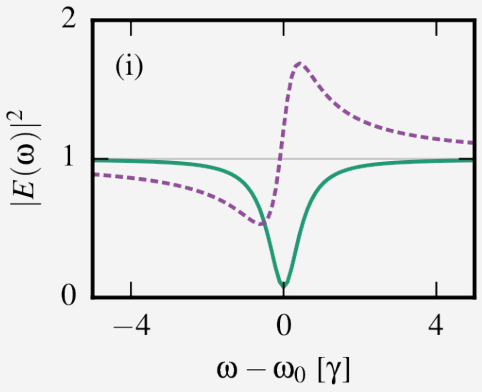
Detect Rabi flopping via spectral interference

Weak x-ray pulse



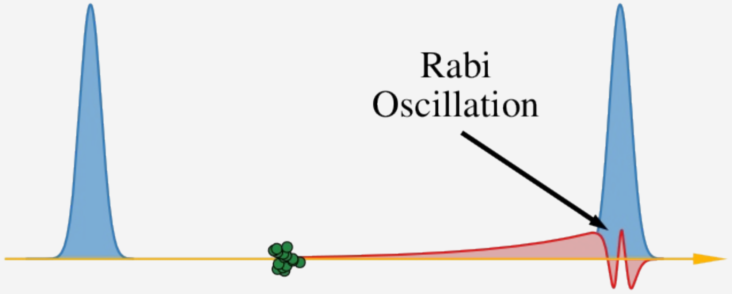


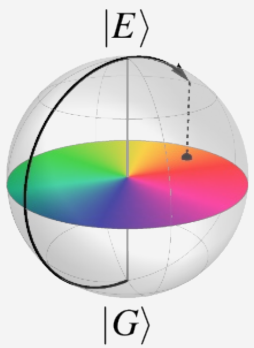
Bloch sphere



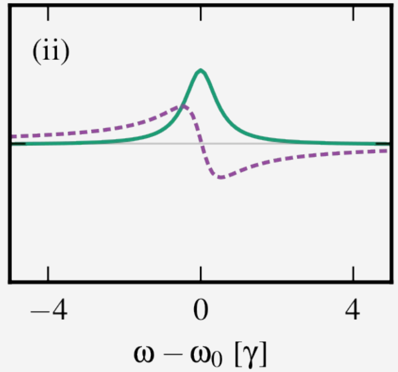
Observed spectrum

Strong x-ray pulse





Bloch sphere



Observed spectrum

Dipole phase change upon each half Rabi cycle leads to flip of spectra

Heeg, Keitel, Evers, in preparation

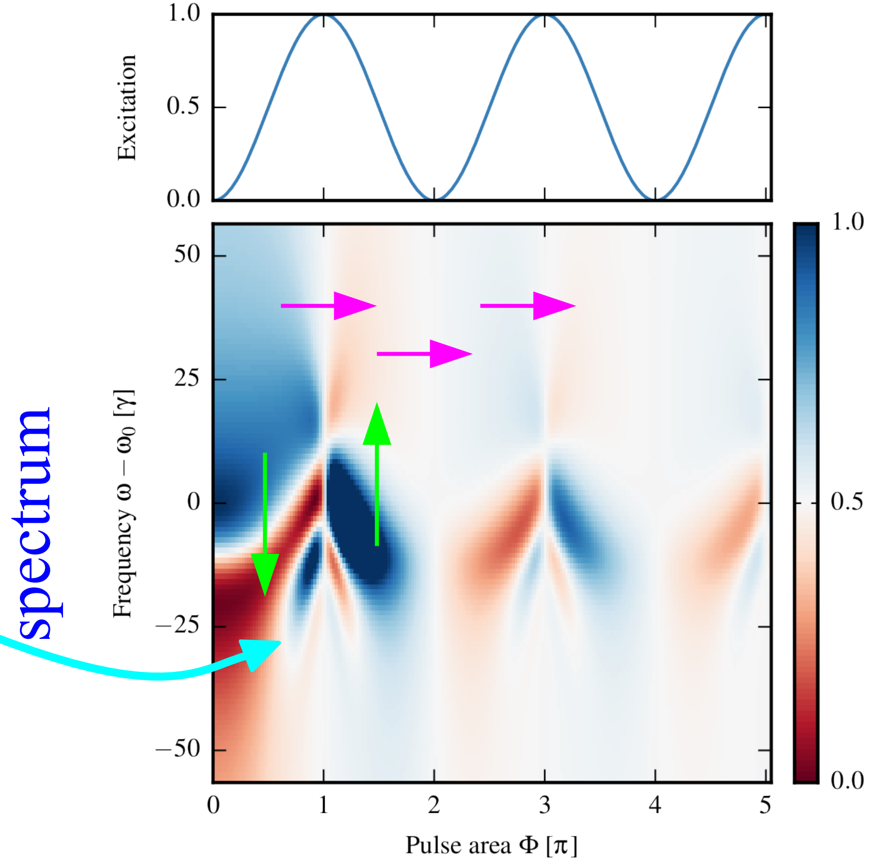
Strongly excited nuclei in x-ray cavities

Approach:

- Extend theory to arbitrary number of excitations in the nuclear ensemble using Dicke model (neglect single particle decay)
- Numerically evaluate for Gaussian and realistic SASE FEL pulses

Result for Gaussian pulse:

- Flip of spectra confirmed
- Additional collective Lamb shifts
- Additional spectral signatures of Dicke model



pulse area

Heeg, Keitel, Evers, in preparation

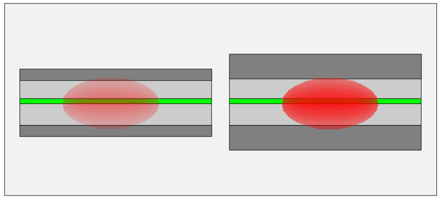
Result for FEL pulse:

- Works for “messy” FEL pulses
- Pulse characterization via total intensity is sufficient
- the cleaner the better, can compensate noise with higher pulse energy

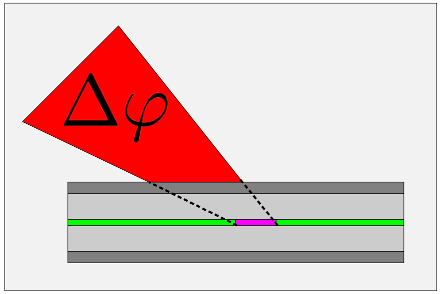
Optimizing the cavity structure

One example: Top layer thickness

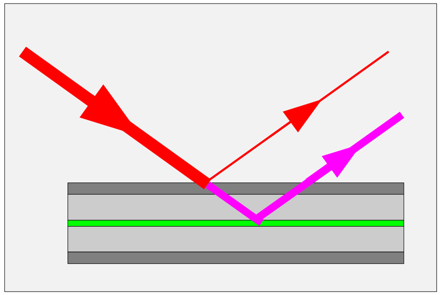
- Thicker layer leads to higher field enhancement
  → Enhanced light-matter interaction



- Thinner layer leads to spectrally broader cavity modes
  → higher angular acceptance of cavity mode
  → stronger focusing possible
  → lower number of nuclei in excitation volume



- Thickness controls visibility of interference between “free” and “scattered” part
  → intermediate thickness favorable

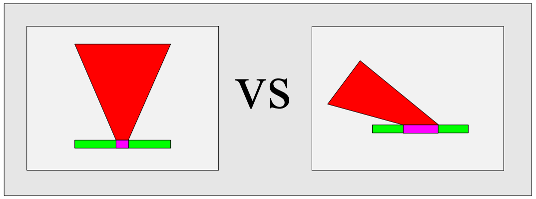


Numerical optimization required

Role of the focusing in cavities

Feasibility of inversion crucially
depends on number of involved nuclei

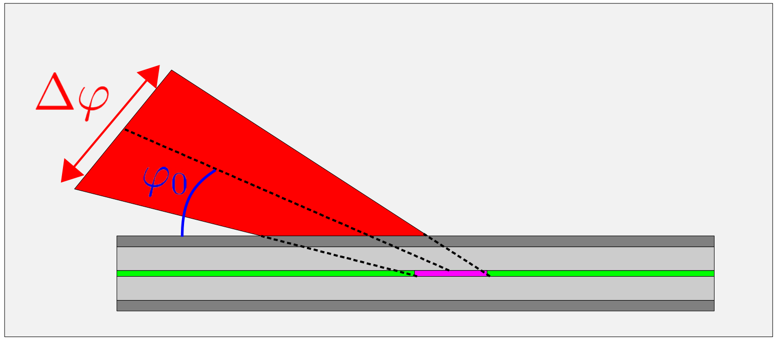
- Thin target layer → less nuclei in target volume
- Grazing incidence enlarges illumination spot (in 1d)
  → more nuclei in target volume



- Focusing reduces of excitation spot (in 2d)
- Angular divergence translates into cavity detuning
  → broadening of x-ray pulse in time domain

$$\Delta_c \approx -\omega \cdot \varphi_0 \cdot \Delta\varphi$$

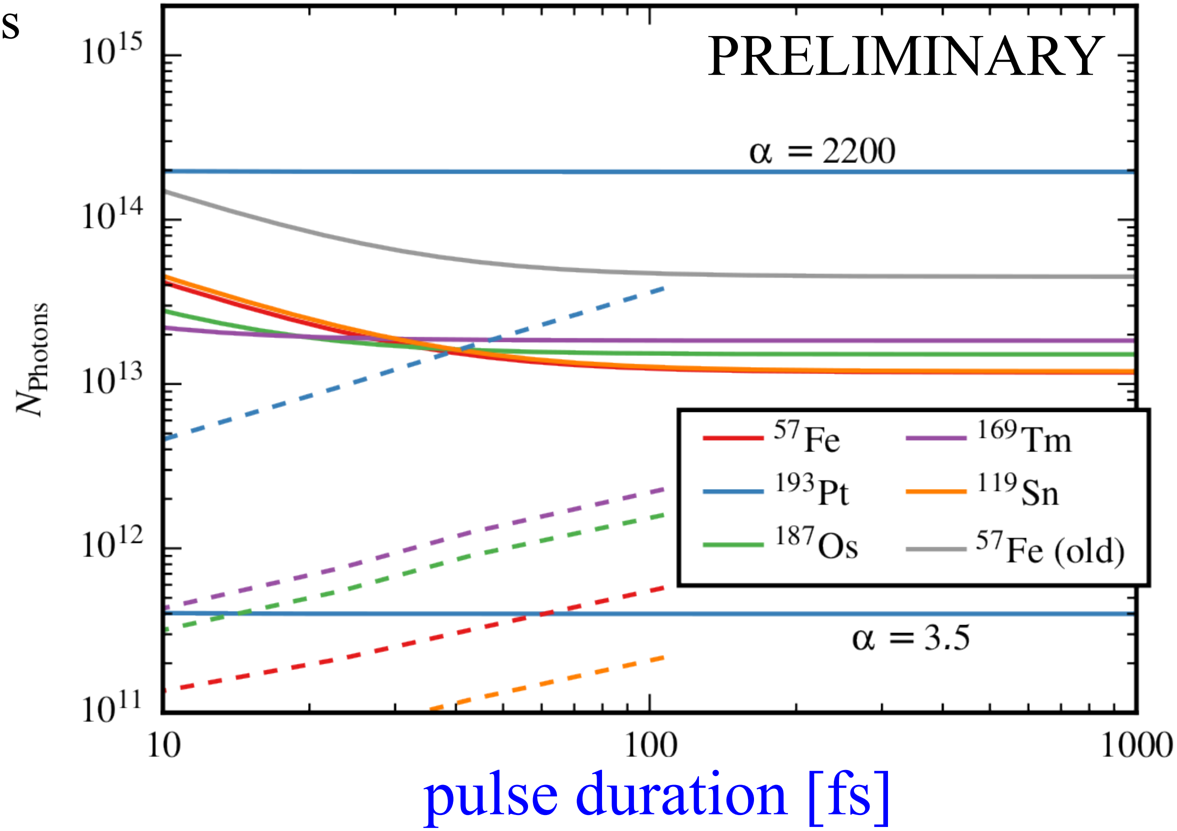
- Focusing limits:
  - * Broadening up to cavity spectral width?
  - * Up to half distance to next mode?
  - * Breakdown of forward scattering?
  - * damage threshold?



Quantitative results

- Temporal pulse broadening due to beam divergence limits advantages of long pulses
- Numbers too high for XFELO, seeded XFEL better ?!?
- Advantage for XFELO: Longer pulse length allows to relax focus in propagation direction
- Note for ^{193}Pt:
  - * $E_0 \sim 1.642$ keV
  - * Unknown $1 \leq \alpha \leq 10^4$
  - * Ground state lifetime 50yr (EC)

Photons required for full inversion



Dashed lines:
TR-2011 SASE photon beam predictions of XFEL

Heeg, Keitel, Evers, in preparation

Summary

- Higher excitation of nuclear ensembles is desirable
(some proposed setups require inversion / Rabi flopping)
- Optimization of target so far typically give 10^1-10^3 reduction of required photon number compared to reported “foil/slab” results
- Probably some further improvement with target optimization possible
- But remember 2 cases of interest:
  * beyond single-photon / single-excitation physics (clearly possible)
  * full inversion (maybe)
- What can be done with few excitations, but not with one?