Prospects of stimulated x-ray Raman scattering with Free-Electron Laser Sources

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Nina Rohringer

Max Planck Institut für Physik Komplexer Systeme, Dresden

Center for Free-Electron Laser Science, Hamburg

CFEL SCIENCE

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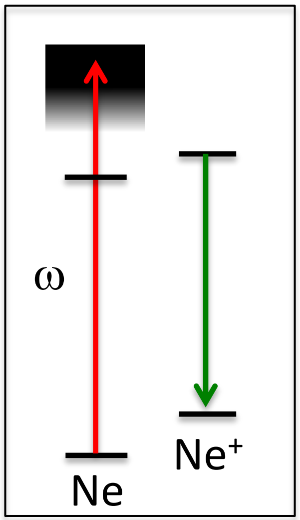
J. Nordgren, J. E. Rubensson

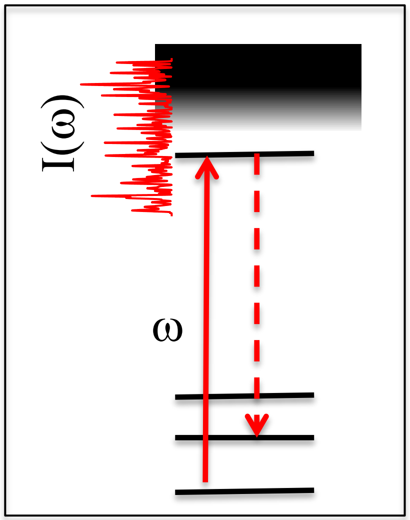
A route to nonlinear spectroscopy with x-rays

Photoionization atomic inner-shell x-ray laser



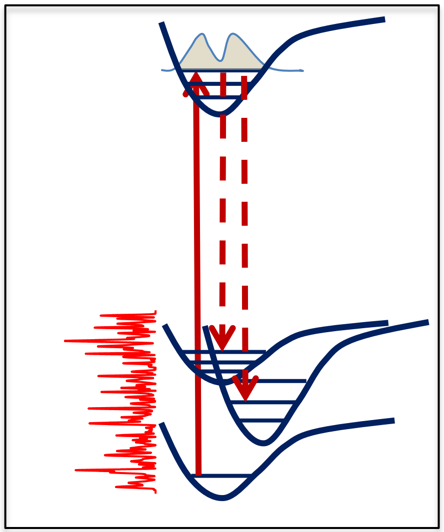
Rohringer et al.,
Nature **481**, 488 (2012)

Stimulated x-ray Raman scattering in atoms



Weninger et al.,
PRL **111**, 233902 (2013)

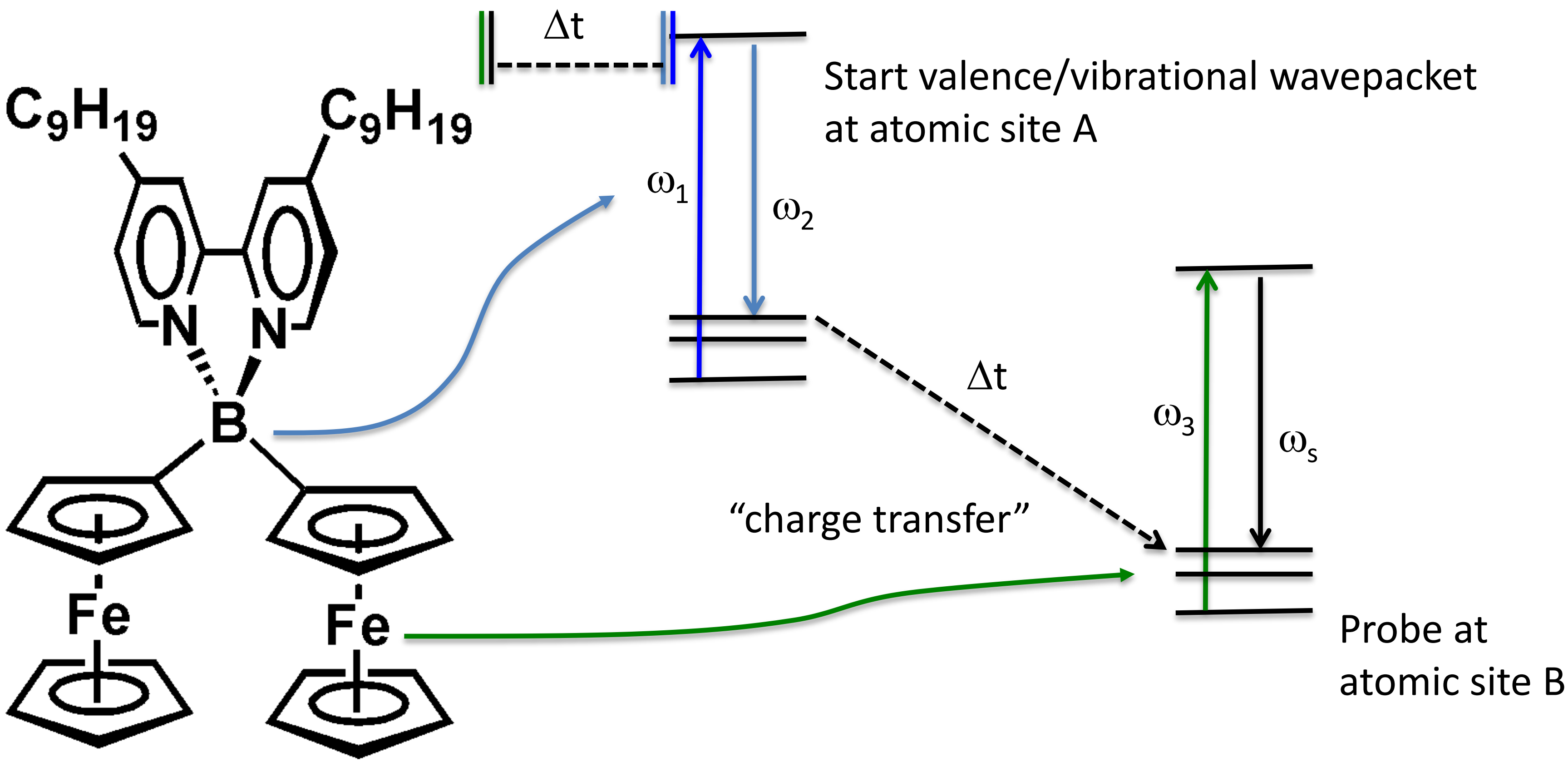
Stimulated x-Raman scattering in molecules



Kimberg & Rohringer,
PRL **110**, 043901 (2012)

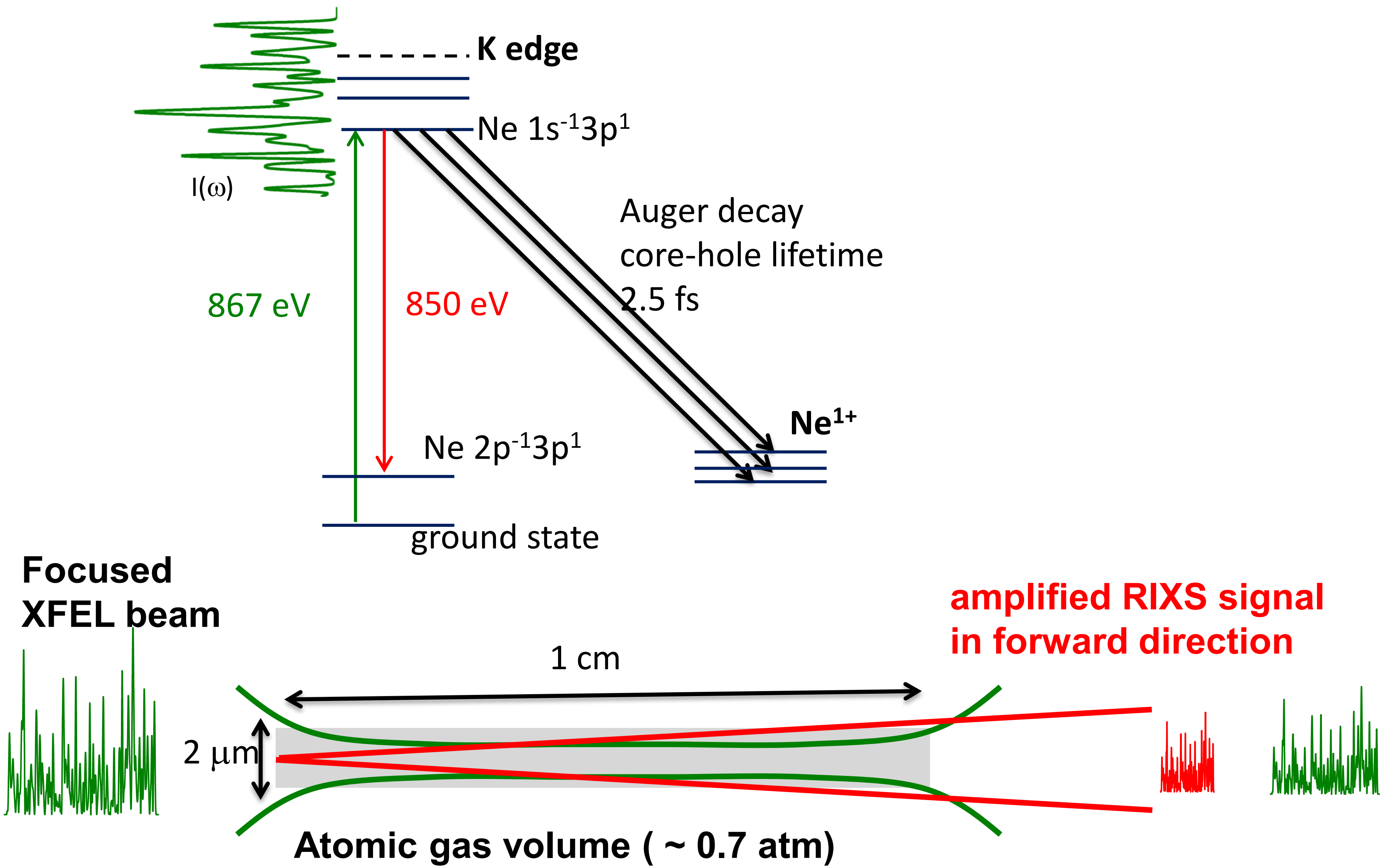
Recent experiment:
LCLS, Feb. 2014
Upcoming experiment:
FLASH, Oct. 2014

Stimulated X-ray Raman scattering
a building block for nonlinear x-ray spectroscopy

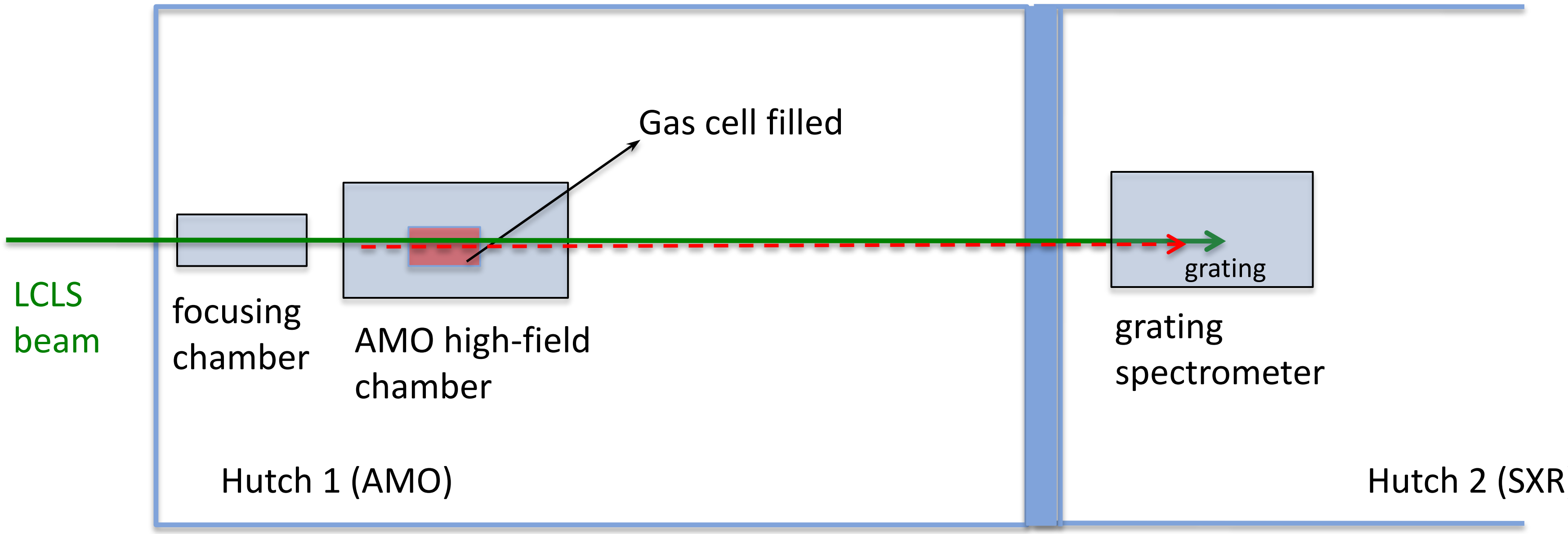


S. Mukamel et al. (PRL 89, 043001 (2002), PRB 72, 235110 (2005); PRA 76, 012504 (2007); PRB 79, 085108 (2009)

Stimulated resonant inelastic x-ray scattering in optically dense gas samples



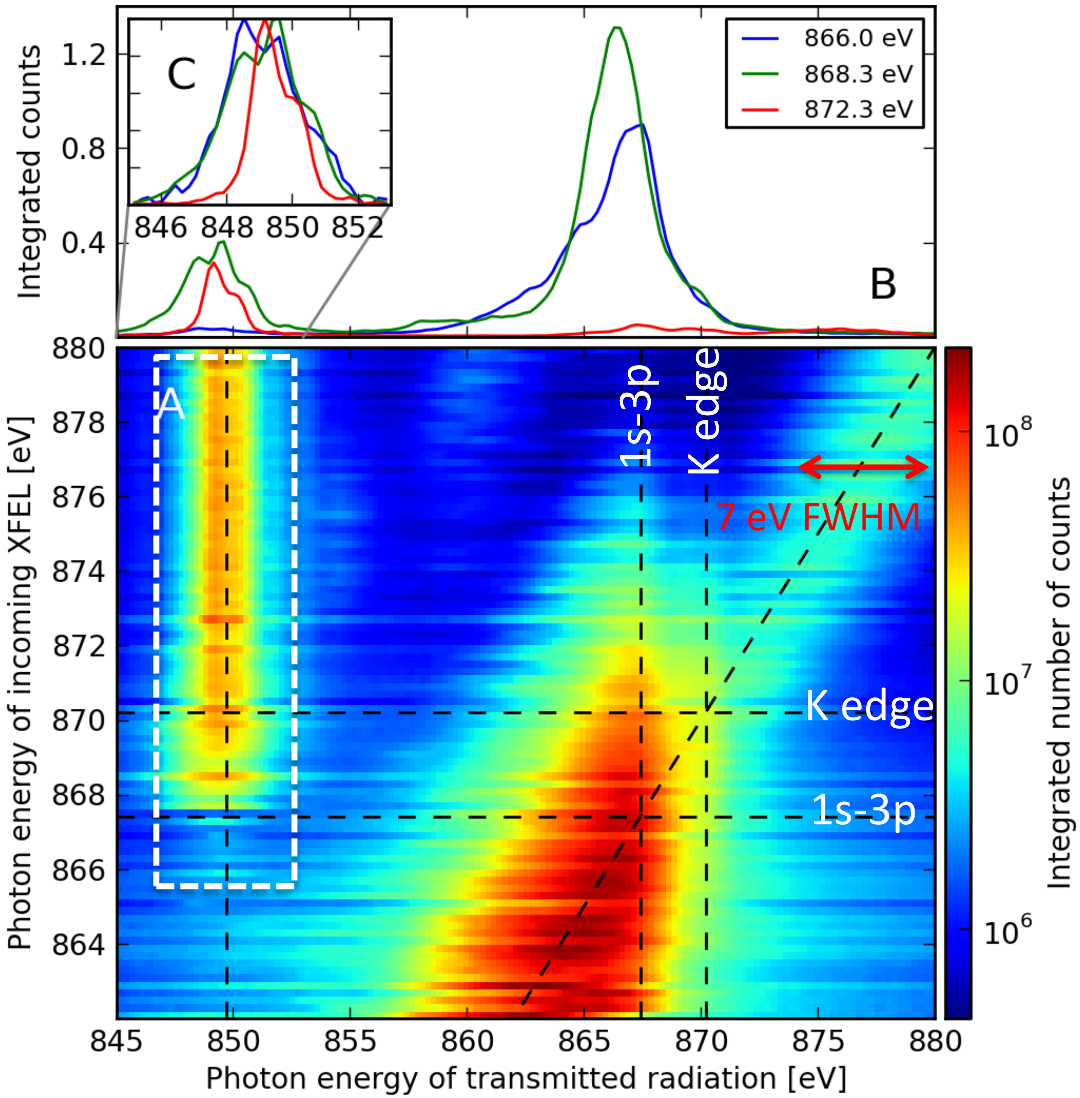
Schematic experimental setup

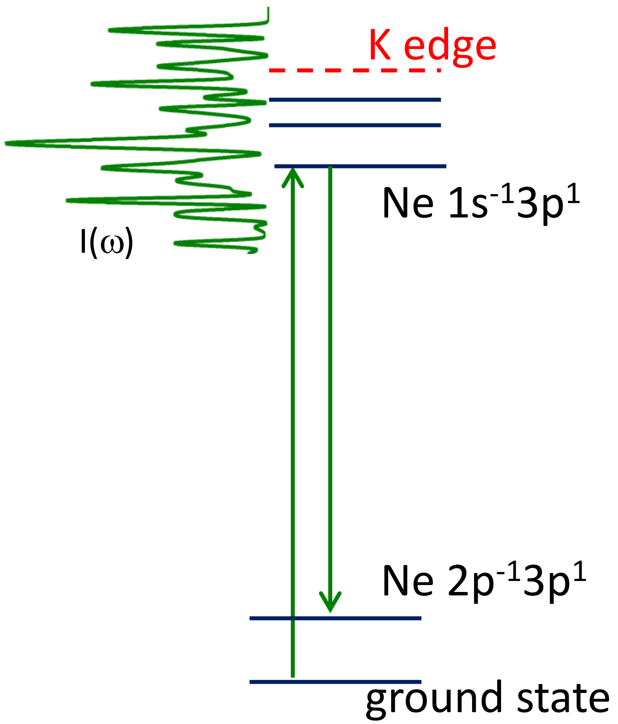


Diagnostics:

- Inline spectrometer for monitoring transmitted XFEL and amplified scattered x rays

1st demonstration of stimulated elecronic x-ray Raman scattering stimulated resonant inelastic x-ray scattering in Neon

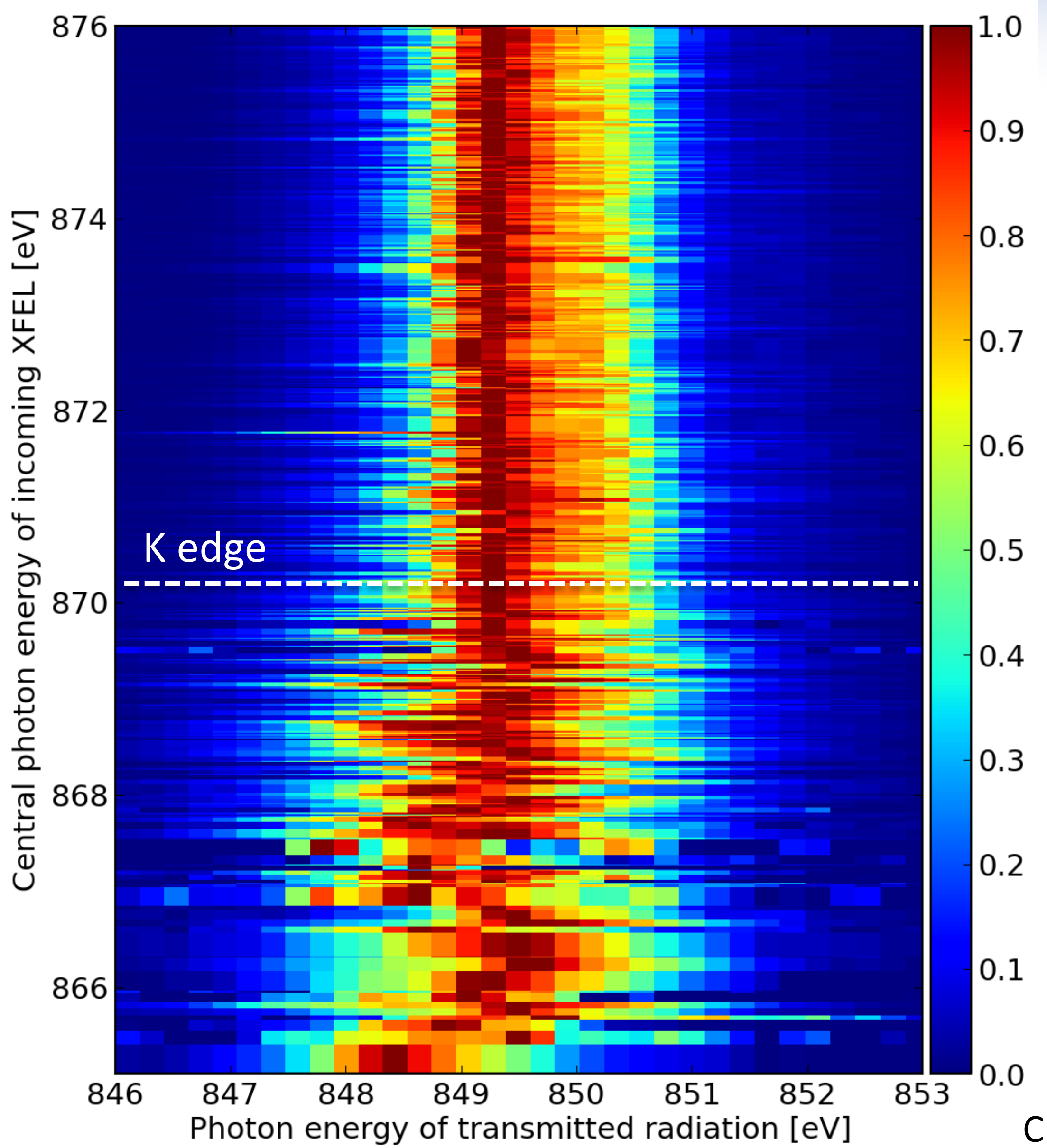




Rohringer et al., Nature **481**, 488 (2012)

Weninger et al., PRL **111**, 233902 (2013)

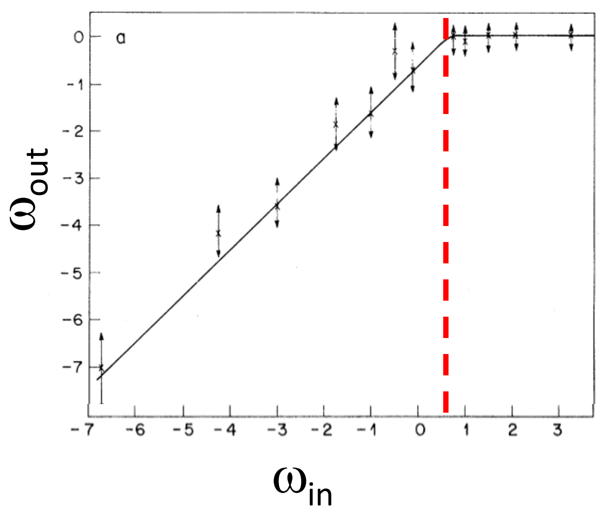
Emitted line profile as a function of pump photon energy



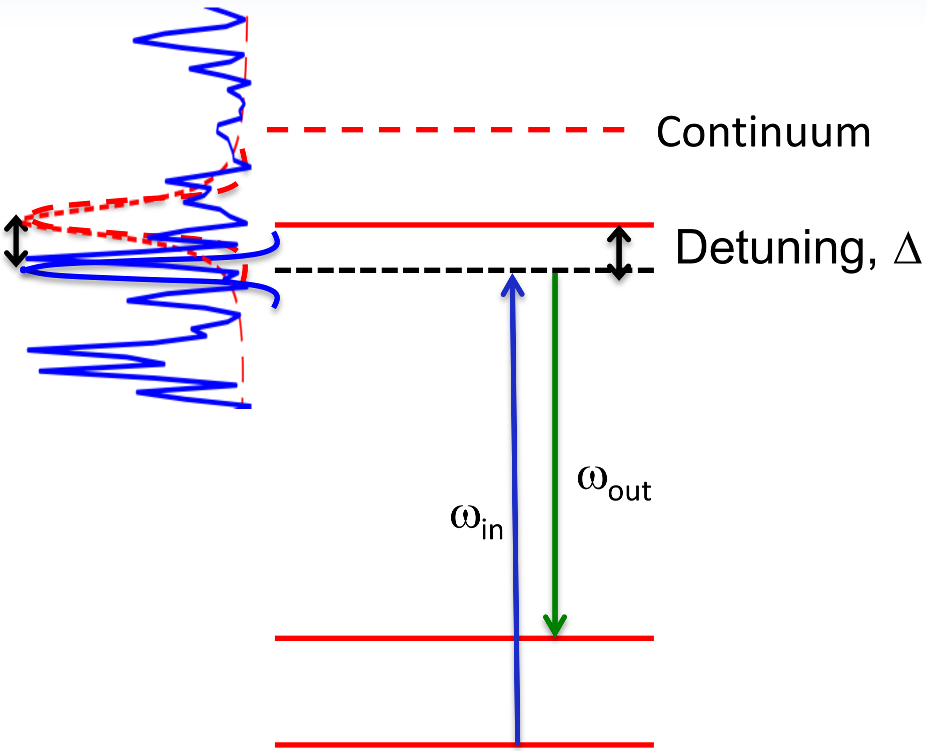
C. Weninger et al.,
Phys. Rev. Lett. **111**, 233902 (2013)

Stochastic line shift due to "anomalous" linear dispersion of resonance scattering

1st RIXS experiments on Cu with Synchrotron radiation (1976)



P. Eisenberger, P.M. Platzman, H. Winick, PRL 36, 623 (1976).

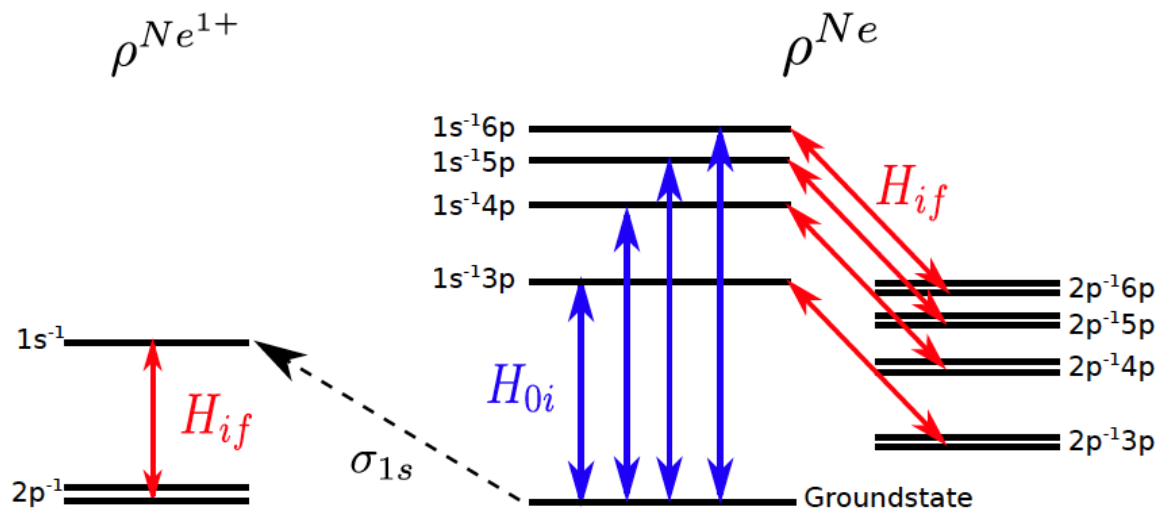


Width of resonance: 0.25 eV

Width of SASE spike: $\Delta\omega=1/\tau$ =0.1 eV

$$\frac{d^2\sigma}{d\Omega d\hbar\omega_2} = r_0^2\left(\frac{\omega_2}{\omega_1}\right)\sum_f \left|\left(\frac{\hbar}{m}\right)\sum_i \sum_{jj'}\left[\frac{\langle f|(\boldsymbol{\epsilon}_2^*\cdot\mathbf{p}_j)e^{-i\mathbf{k}_2\cdot\mathbf{r}_j})|i\rangle\langle i|(\boldsymbol{\epsilon}_1\cdot\mathbf{p}_{j'})e^{i\mathbf{k}_1\cdot\mathbf{r}_{j'}})|g\rangle}{E_g - E_i + \hbar\omega_1 - i\Gamma_i/2}\right.\right|^2 \delta(E_g - E_f + \hbar\omega_1 - \hbar\omega_2)$$

Master Equations for atomic and ionic density matrices coupled to Maxwell's equation



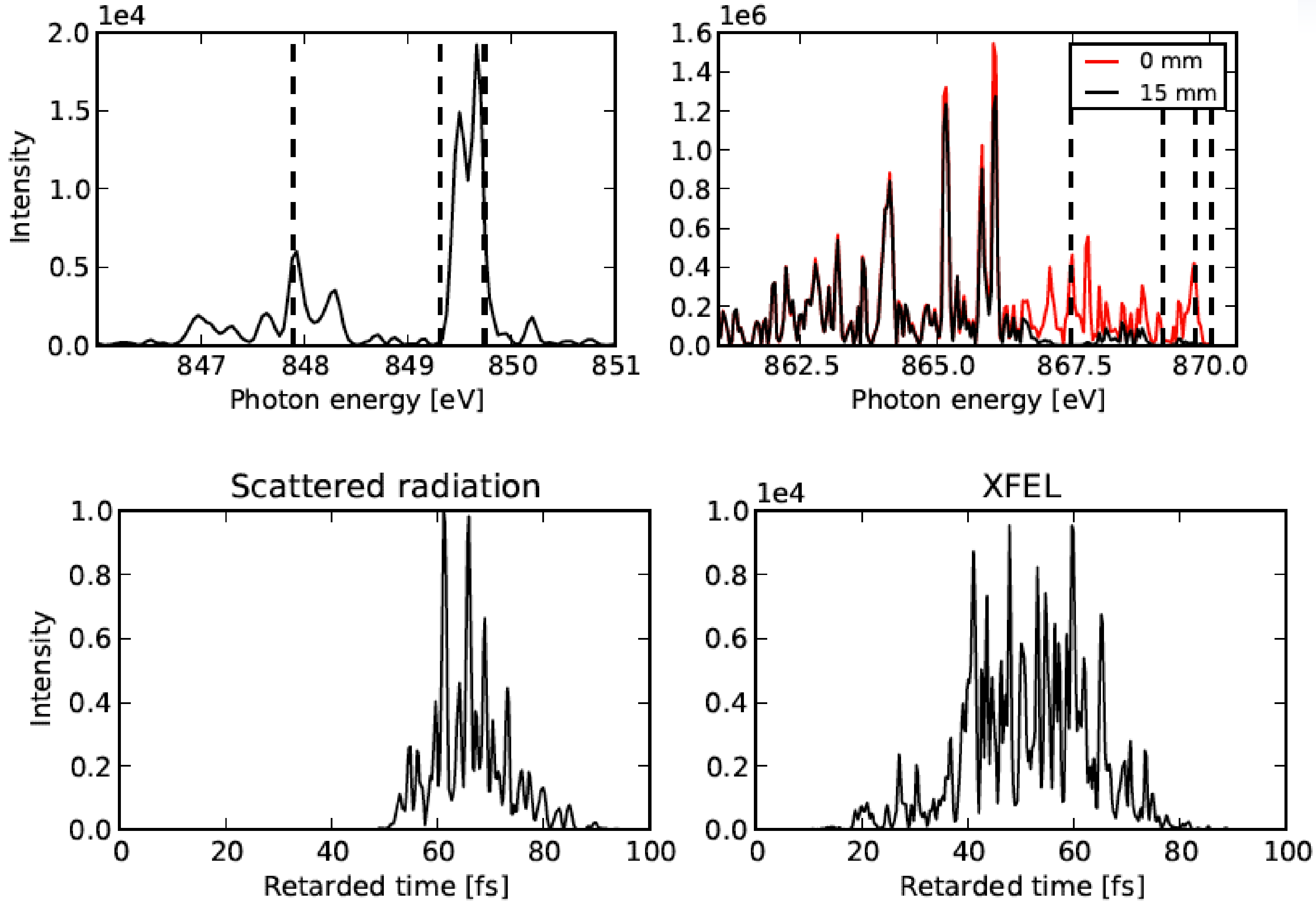
$$\frac{\partial}{\partial t}\hat{\rho} = -i[\hat{H},\hat{\rho}] - \hat{\Gamma}\hat{\rho} - \sigma\hat{\rho}I_p + S$$

$$\mathscr{P} = 2n\left(\sum \mu_{ij}\rho_{ij}^{\mathrm{Ne}^{1+}} + \sum \mu_{kl}\rho_{kl}^{\mathrm{Ne}}\right)$$

$$\frac{\partial \mathscr{E}(\tau,z)}{\partial z} = i\frac{2\pi\omega}{c}\mathscr{P}(\tau,z)$$

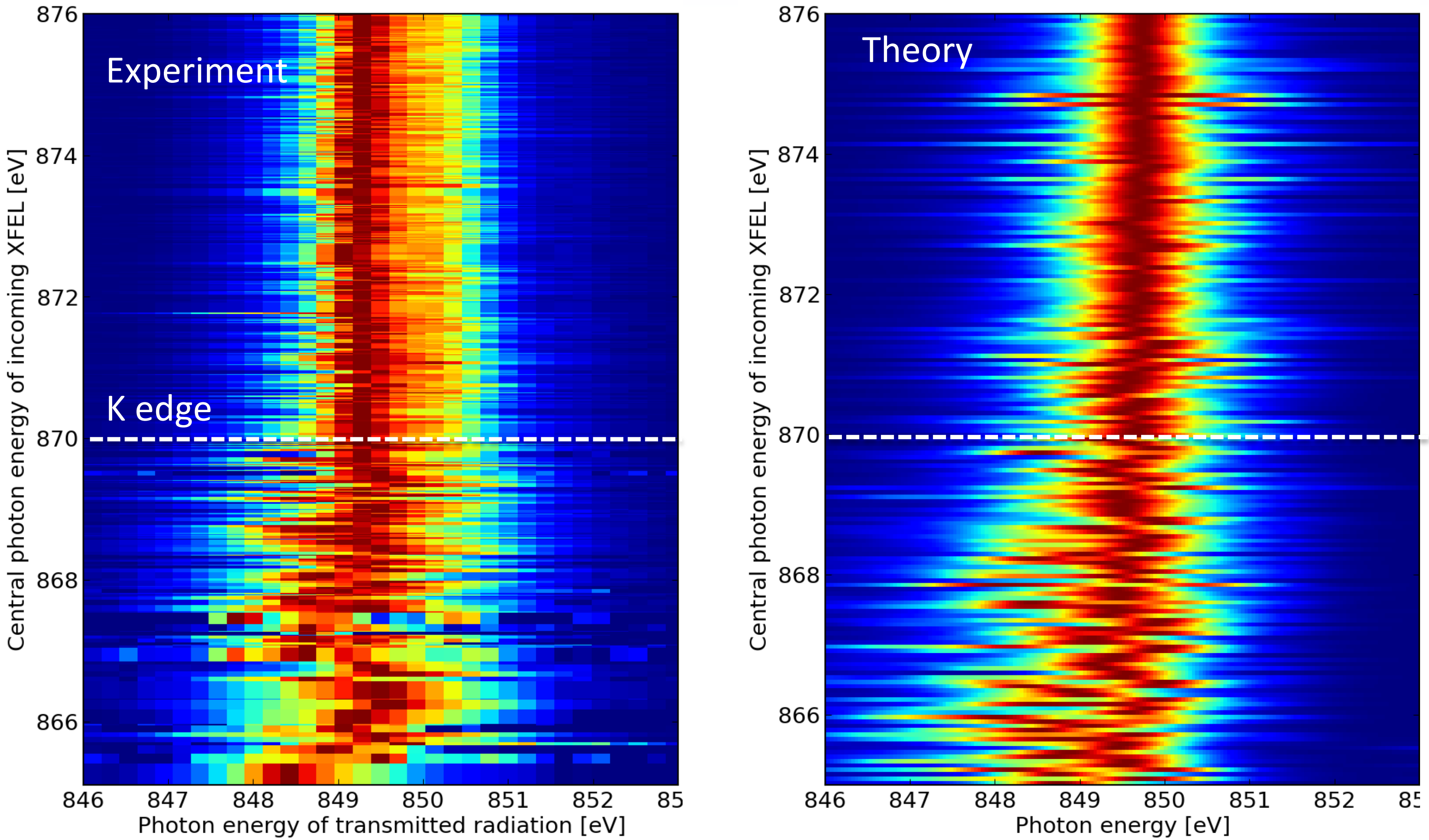
C. Weninger & N.R., Phys. Rev. A **88**, 053421 (2013)

Simulated spectral / temporal intensity profiles of sRIXS process



C. Weninger & N.R., Phys. Rev. A **88**, 053421 (2013)

Line profile – comparison of experiment to simulation

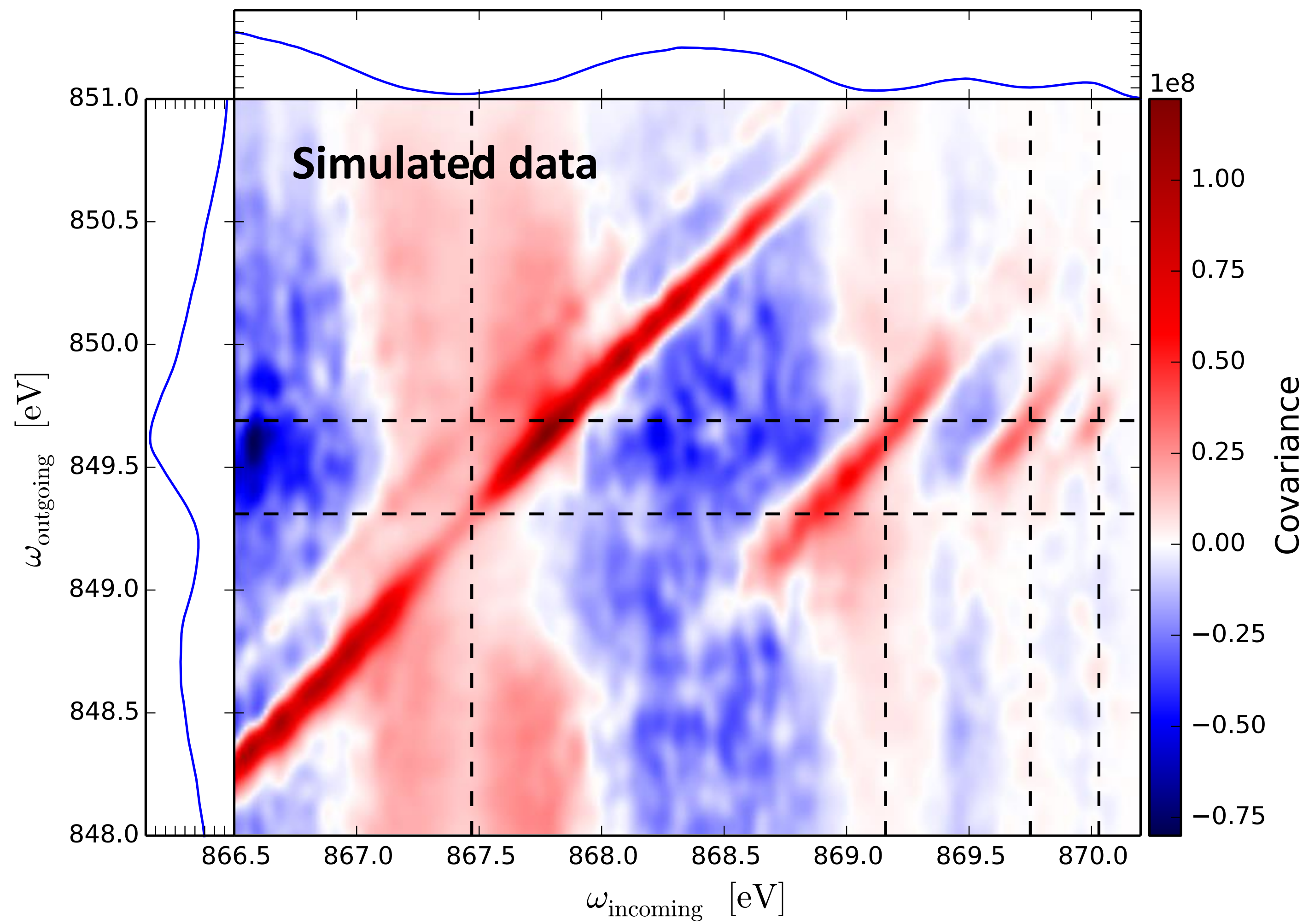


C. Weninger et al.,
Phys. Rev. Lett. **111**, 233902 (2013)

High-resolution x-ray Raman spectroscopy by statistical analysis (covariance mapping)

$$Cov(\omega_1,\omega_2) = \langle I(\omega_1)I(\omega_2)\rangle - \langle I(\omega_1)\rangle\langle I(\omega_2)\rangle$$

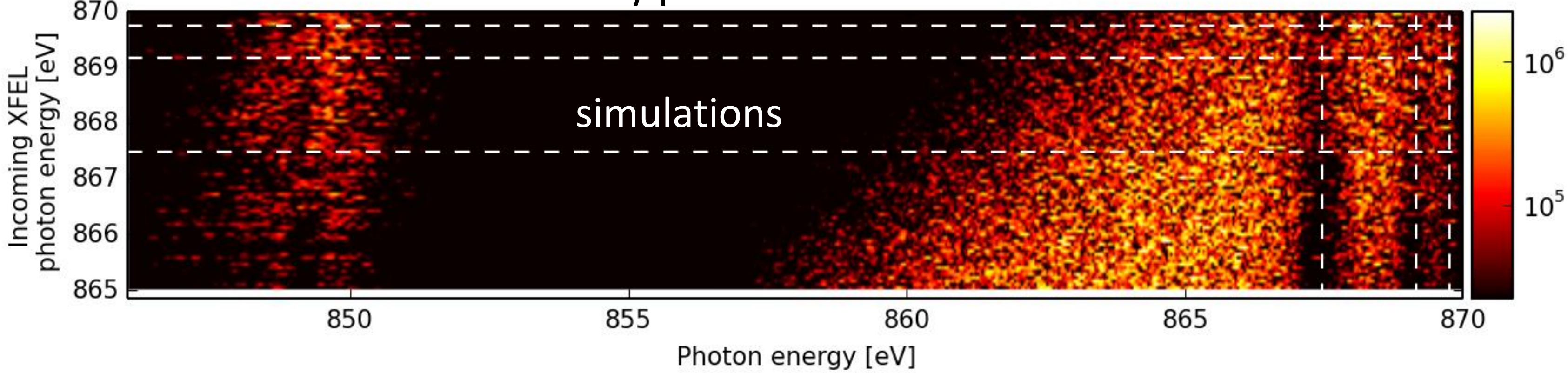
Covariance map from 5000 simulated single-shot



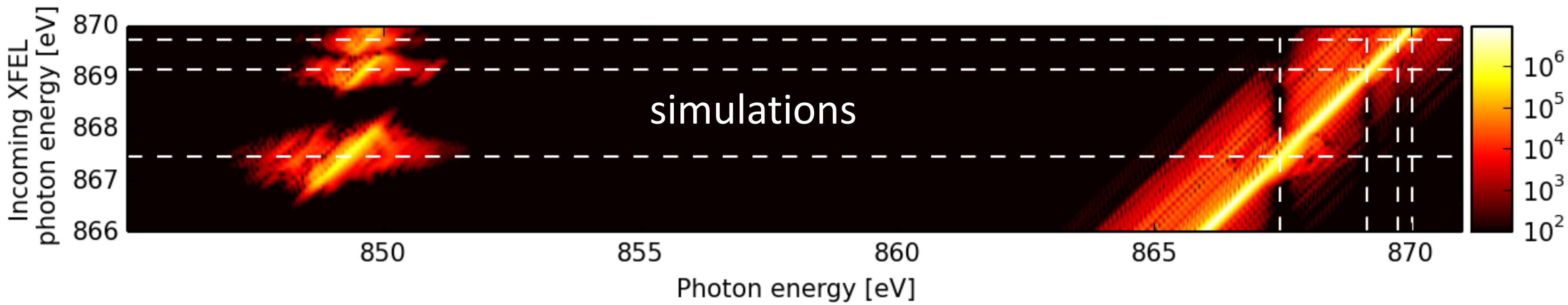
C. Weninger & N.R., Phys. Rev. A **88**, 053421 (2013)

Soft x-ray laser seeding at LCLS – new opportunities for stimulated Raman scattering

SASE x-ray pulse

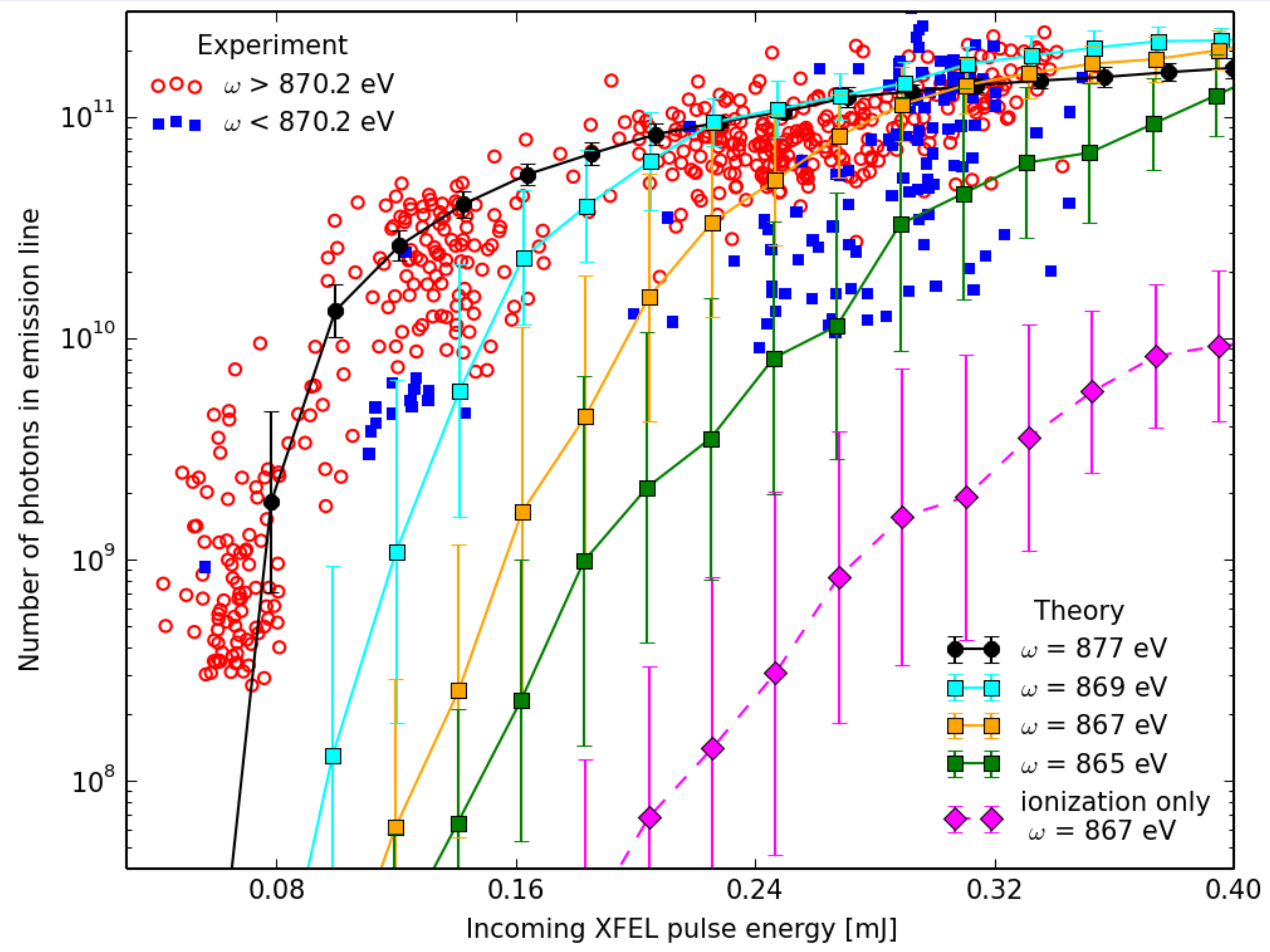


Self-seeded x-ray pulse, with SASE background



2-color mode within SASE bandwidth possible: 1 color self-seeded, 2nd color SASE
Short pulses possible by using slotted foil

Raman Signal Strength as a Function of Pump Energy

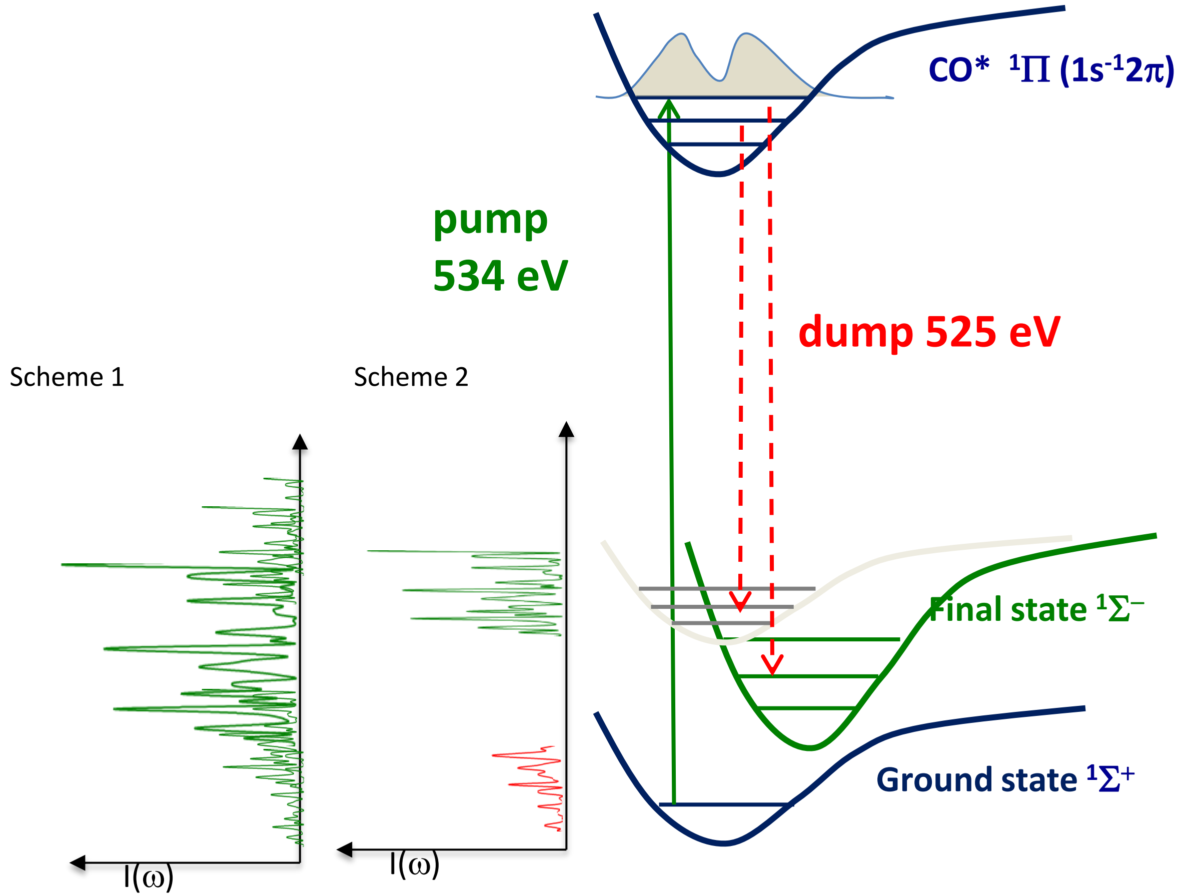


Number of seed photons: 10^3-10^4 (varying due to spectral sidebands of the XFEL)
Estimated number of photons of spontaneous RIXS: 100
Saturated amplification by 7-8 orders of magnitude

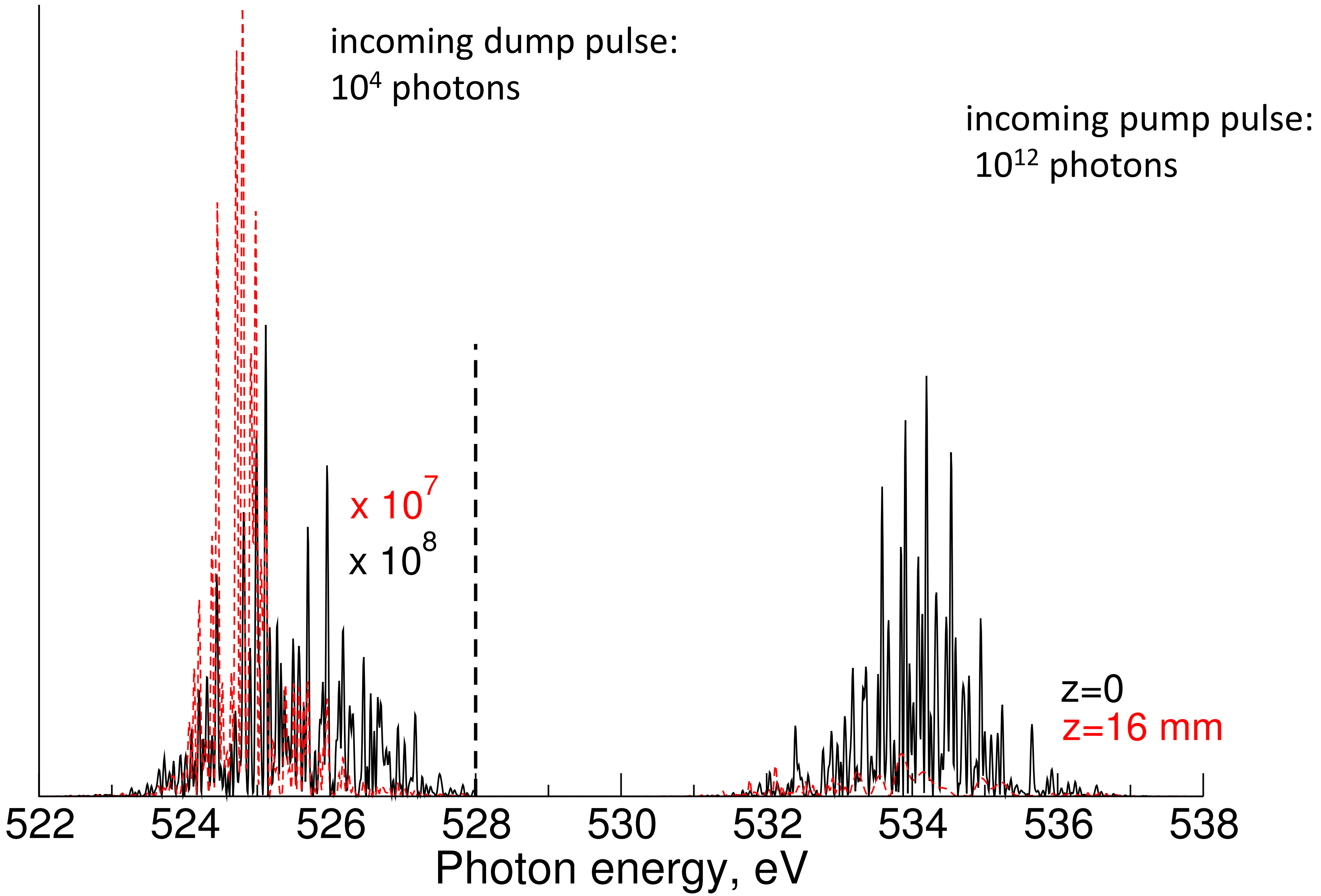
C. Weninger et al.,
Phys. Rev. Lett. **111**, 233902 (2013)

Results of recent experiment at the AMO instrument @ LCLS

February 6-10, 2014 (60 hours of beamtime)

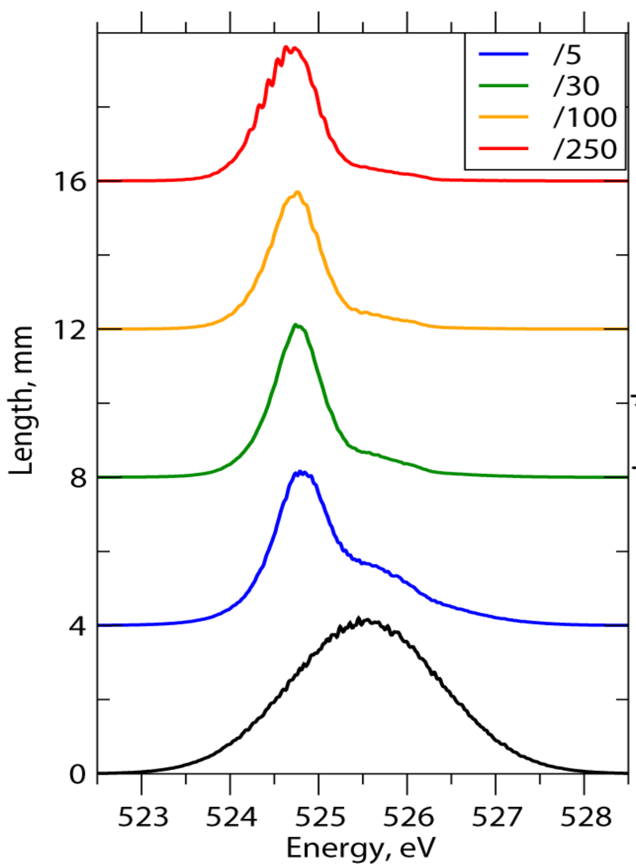


Simulated RIXS spectra on π* resonance in CO strong pump pulse

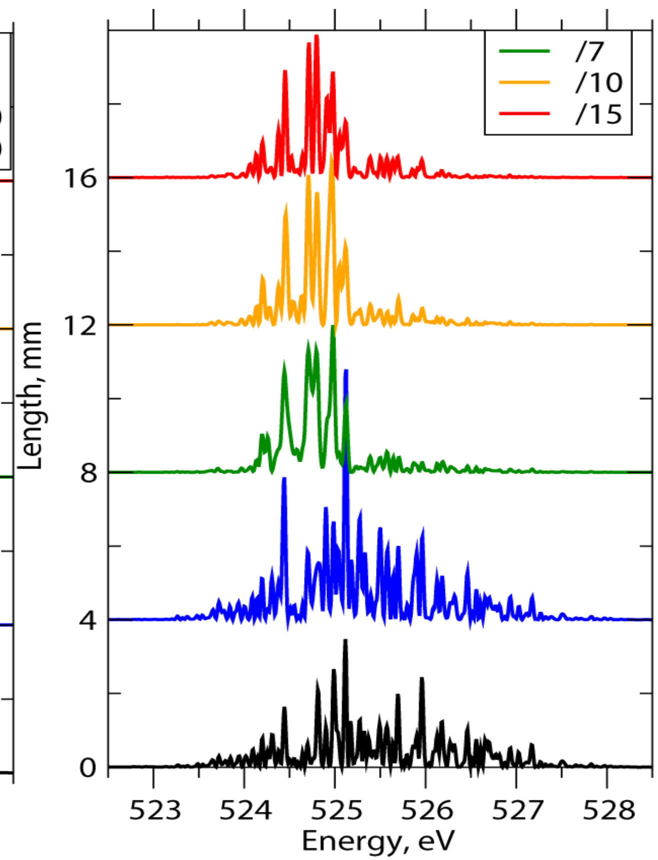


Predicted emission spectrum as a function of gain-length strong pumping regime

Averaged over 5000 shots



Single SASE shot



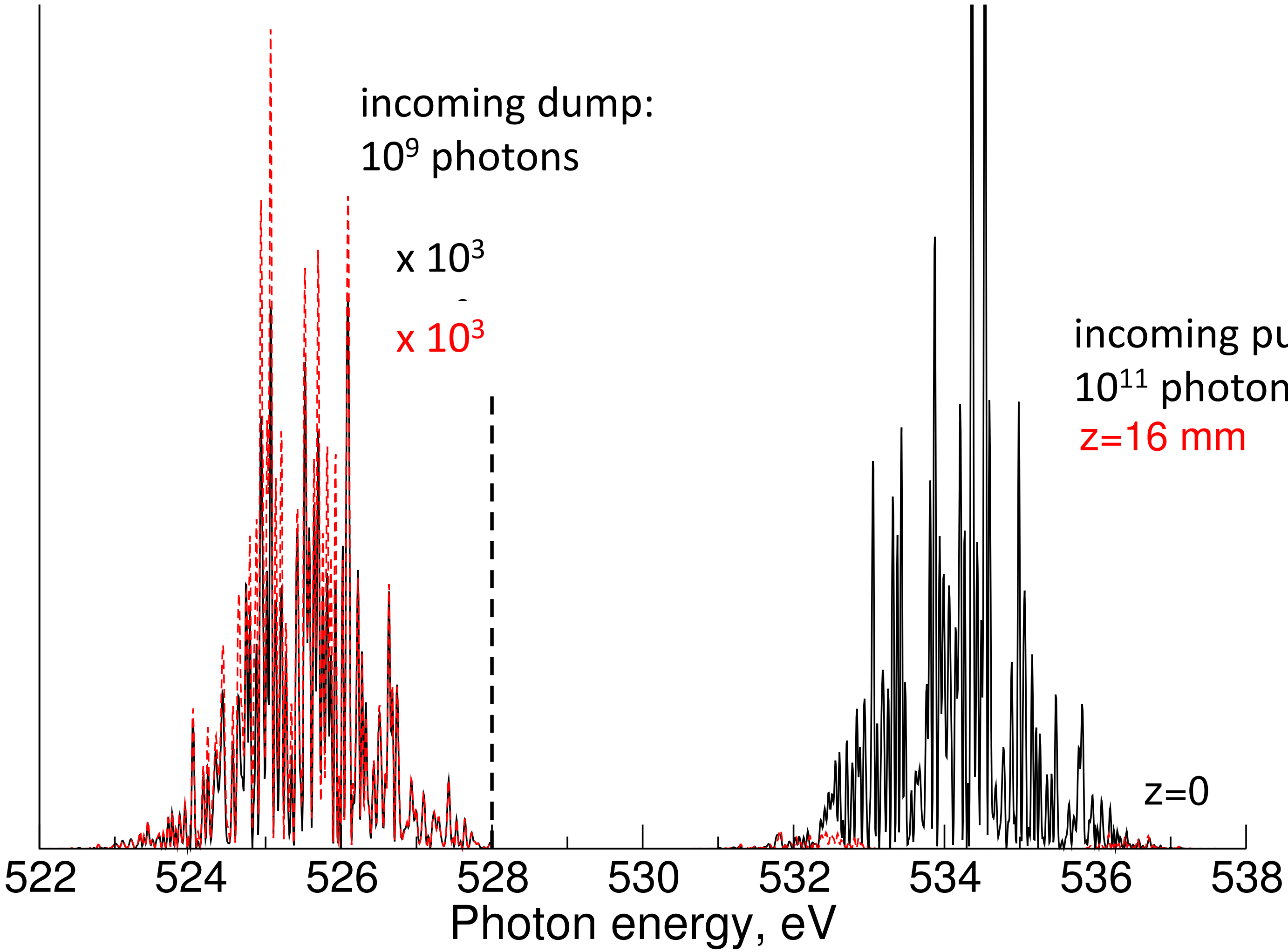
- Strong pump field propagation effects
- Rabi splitting of the core-excited transitions
- Induced ground state wave packet dynamics
- Non-linear propagation effects

Kimberg & Rohringer, manuscript in preparation

Simulated RIXS spectra on π* resonance in CO weak pump pulse

Challenge: Find regime of best signal contrast (at least ~20-30% change)

Raman gain has to statistically exceed SASE fluctuation

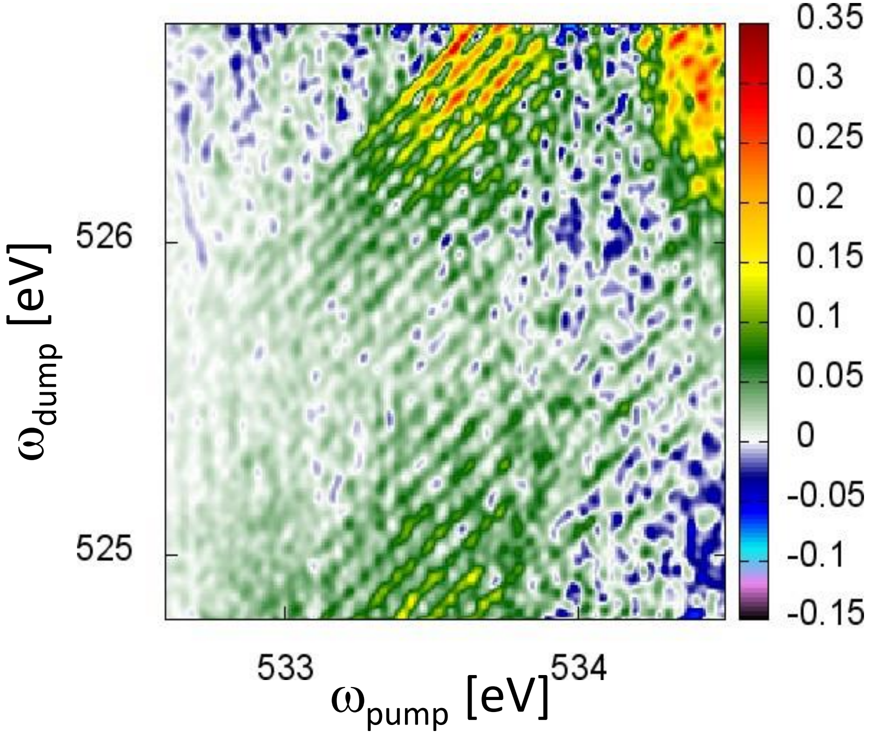


Kimberg & Rohringer, manuscript in preparation

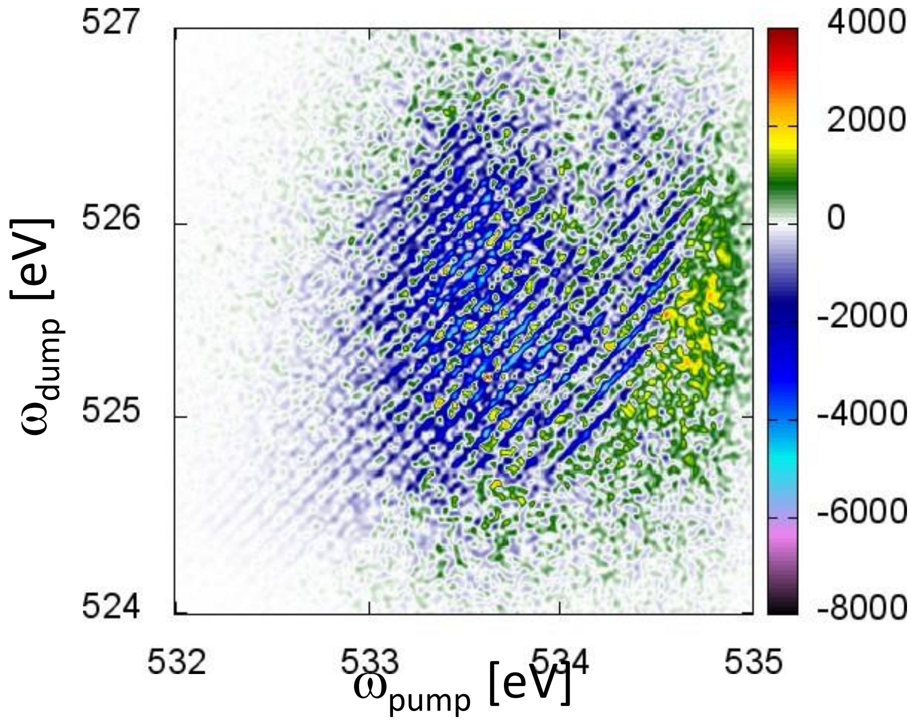
Covariance map of 5000 simulated single-shot spectra

- $N_{pump}=N_{dump}=10^{10}$ photons
- Resolution of the vibrational structure

Covariance map of transmitted spectra

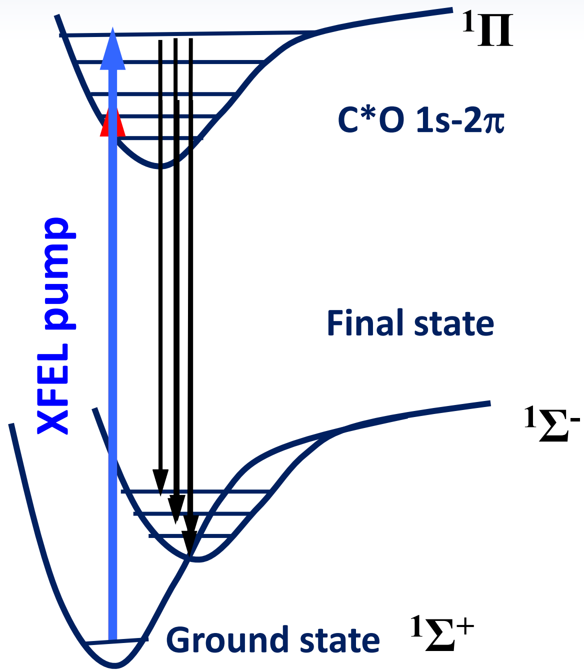


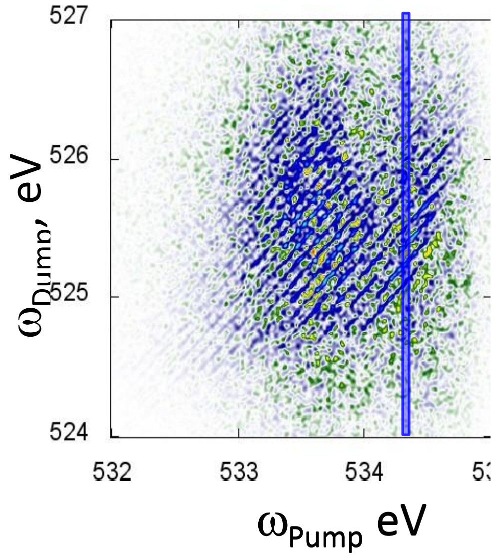
Covariance map of difference spectra
(incoming – outgoing spectrum)

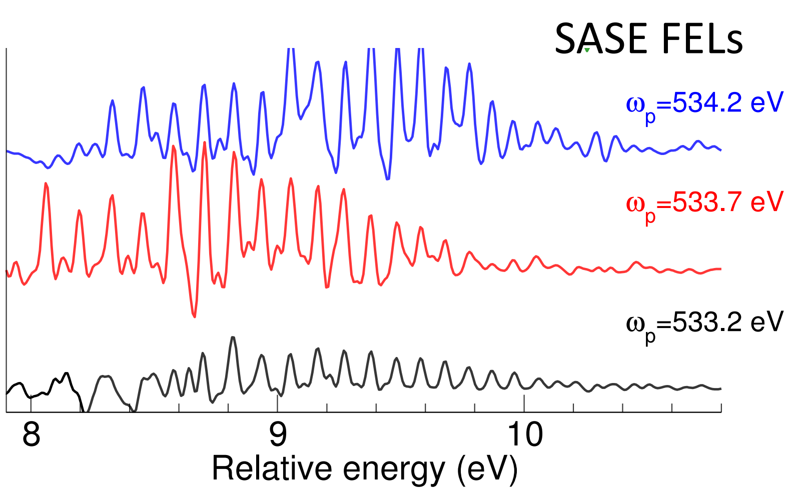


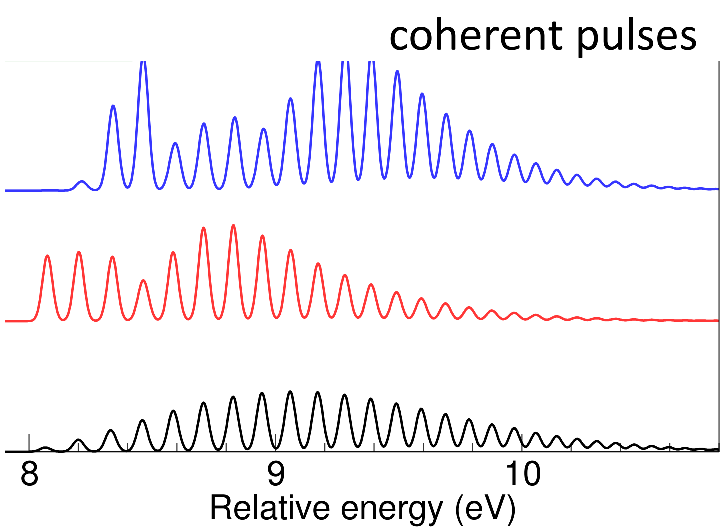
Kimberg & Rohringer, manuscript in preparation

Mapping potential energy surfaces



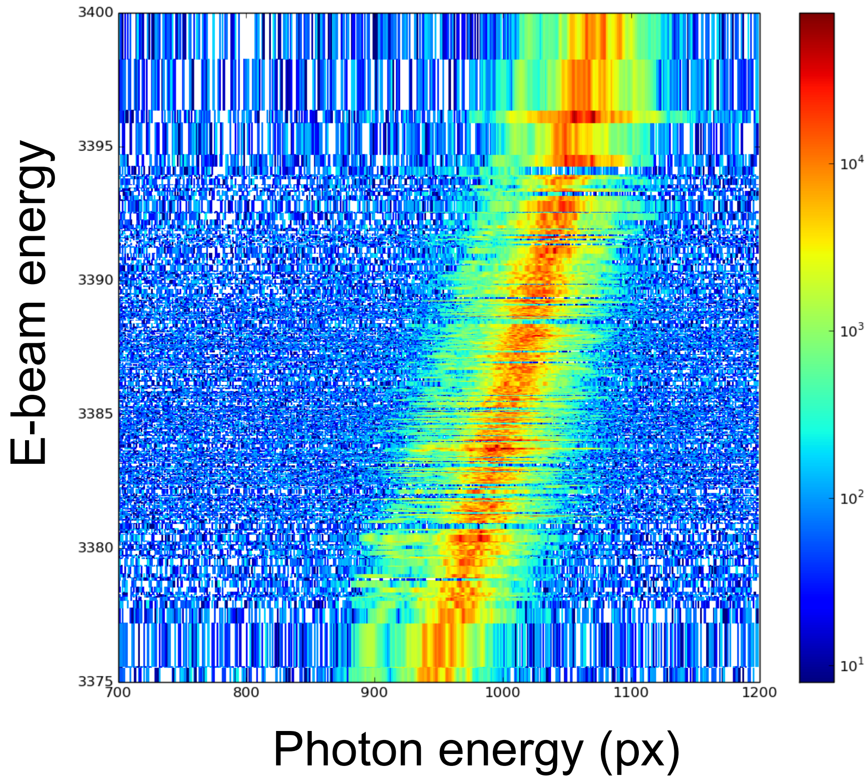




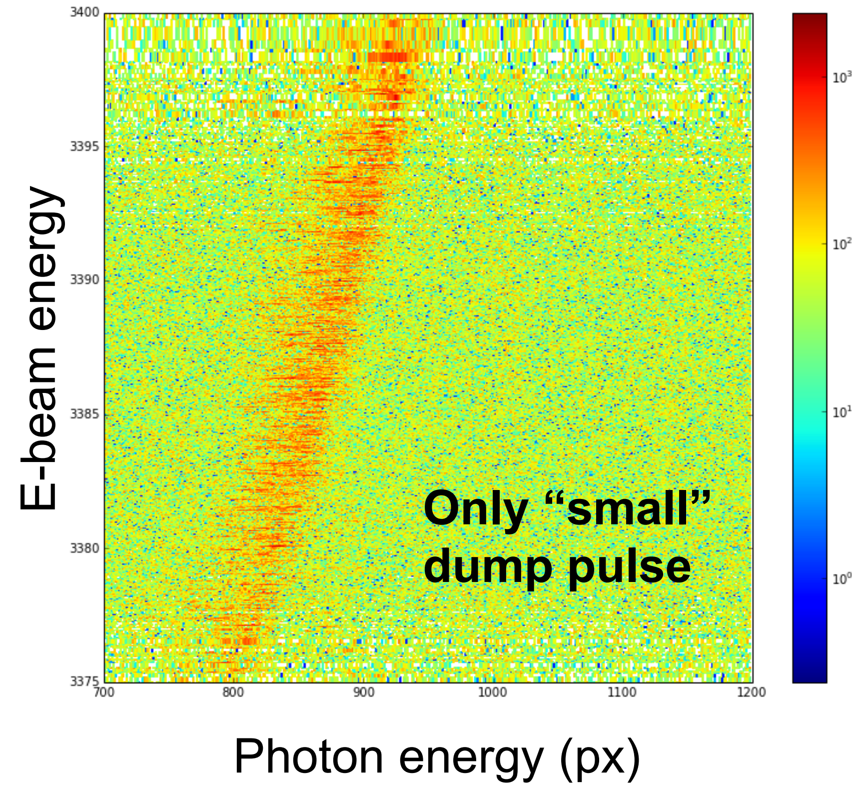


Two-color SASE mode (shift 5, no gas)

534 eV ("big" pulse on pump transition, 0.75 – 1 mJ pulse energy),
525 eV ("small" pulse on dump transition, ~ 0.01 mJ pulse energy)



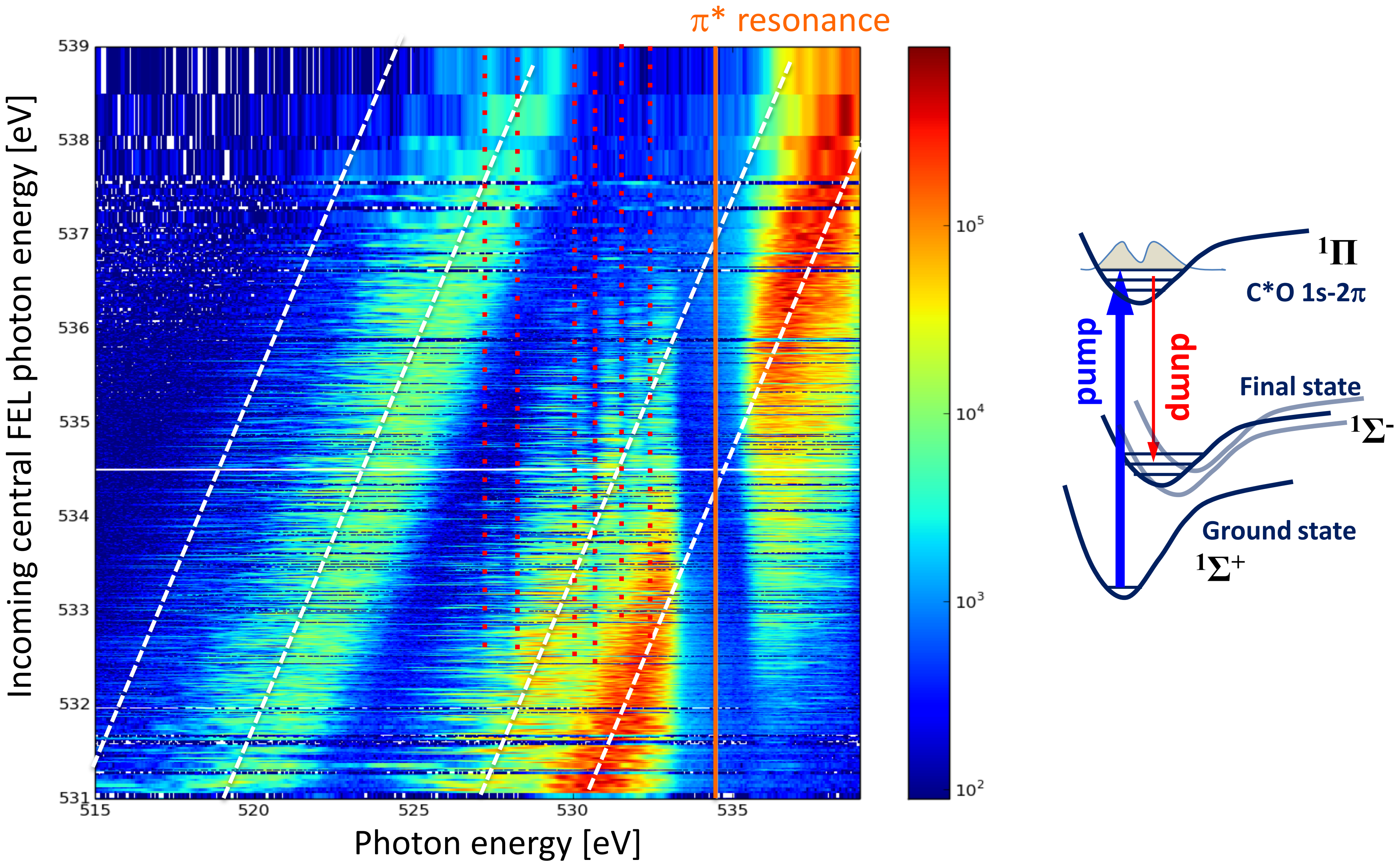
Two colors
("small" dump pulse hardly visible)



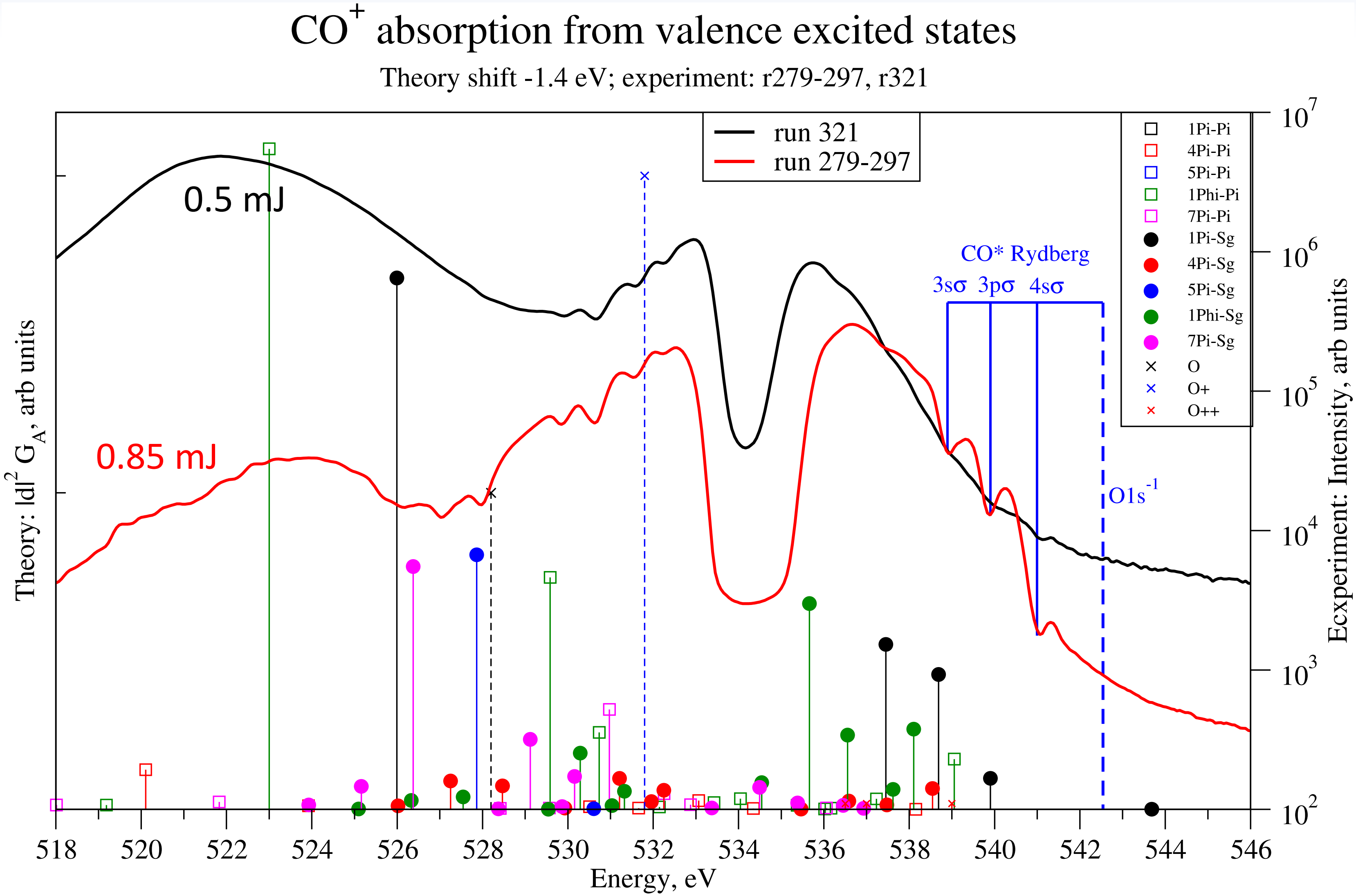
to measure "small" pulse, electron bunch is "kicked out" before the second set of undulators

2-color SASE modes:
Lutman et al., PRL **110**, 134801 (2013).
A. Marinelli et al., PRL **111**, 134801 (2013)

Two-color stimulated RIXS in CO
Spectra as a function of incoming photon energy



Identifying the zoo of absorption and interference (?) dips



Challenges & Outlook

- Large spectral shot-to-shot fluctuation on top of SASE fluctuation (e-beam energy)
  - -> hampers observation of sRIXS in averaged spectra
  - -> requires recording of several 10.000 single-shot spectra

- Assess spectral stability and necessary gain regime for future self-seeded and seeded FELs
  - -> Optically dense samples pose real problems for spectroscopy (pulse propagation, spectral distortions,...)

- Need for fast read-out, small-pixel detectors with high dynamic range

-Model calculations starting from vibrational wave-packets, to probe real dynamics

-Need to develop statistical analysis techniques beyond covariance analysis and link it to higher-order susceptibilities

- Study of coupled nuclear-electron dynamics (vibronic coupling, conical intersections,...)

- New detection schemes beyond simple forward scattering geometry
  - -> transient grating spectroscopy
  - -> non-linear photo- or Auger-electron spectroscopy