Stimulated Electronic X-Ray Raman Scattering in atomic and molecular gases

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Nina Rohringer

CFEL SCIENCE

Max Planck Institute for Structure and Dynamics of Matter,
Center for Free-Electron Laser Science, Hamburg

MAX-PLANCK-GESELLSCHAFT

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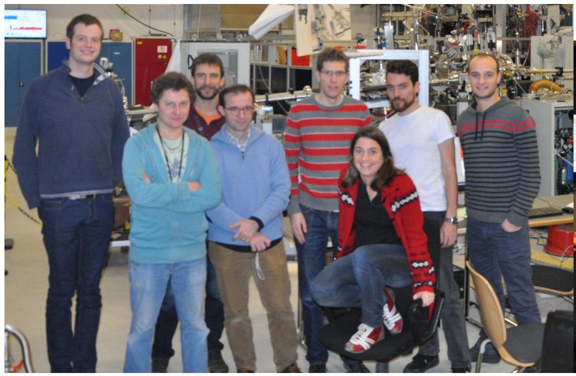
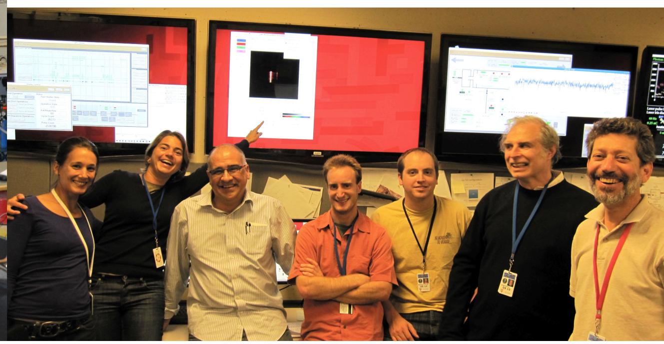
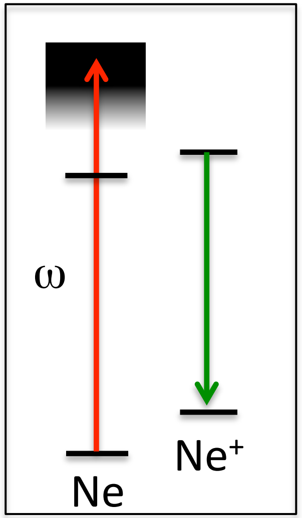




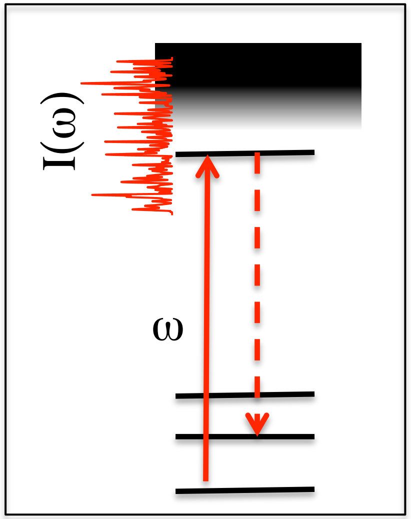
A route to nonlinear spectroscopy with x-rays

Photoionization atomic inner-shell x-ray laser



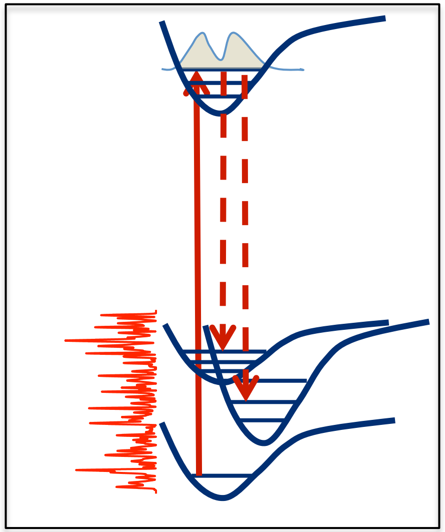
Rohringer et al.,
Nature **481**, 488 (2012)

Stimulated x-ray Raman scattering in atoms



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

X-ray amplification and wave-packet dynamics in molecules

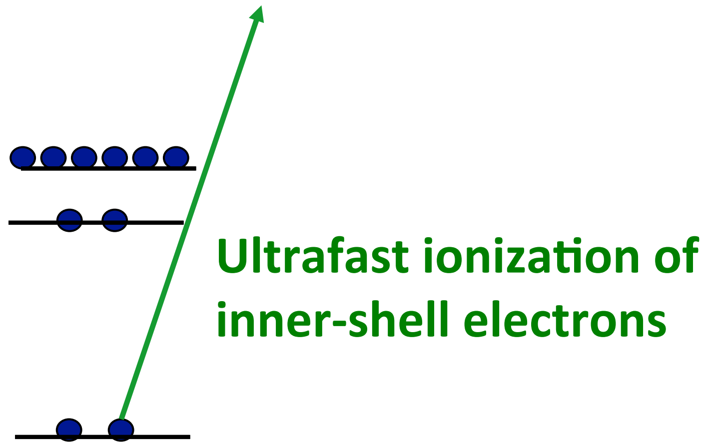


Kimberg & Rohringer,
PRL 110, 043901 (2012)

1^{st} theoretical concept of an atomic X-ray laser
Population inversion by inner-shell photoionization

Duguay and Rentzepis,
Appl. Phys. Lett. 10, 350 (1967).

Realized in the optical regime:
Silfvast et al. 1983 (blue laser)



Fast, powerful x-ray pump required to beat Auger decay !

History of photo-ionization X-ray Lasing schemes

Pump with laser produced x-ray sources

1967 (th.) Duguay and Rentzepis, Appl. Phys. Lett. 10, 350 (Na 33 eV, Cu 9 keV)
1976 (th.) Axelrod (Su)
1983 (exp.) Silfvast et al. 1983 (blue laser)
1992 (th.) Kapteyn, Appl. Opt. 31, 4931 (Ne, 850 eV)
1993-98 (th.) Eder, Strobel, Moon, London, et al.

Use laser-generated betatron source to pump XRL

2007 (th.) Jacquemot, Phuoc, Rousse, Sebban (N, Ne)

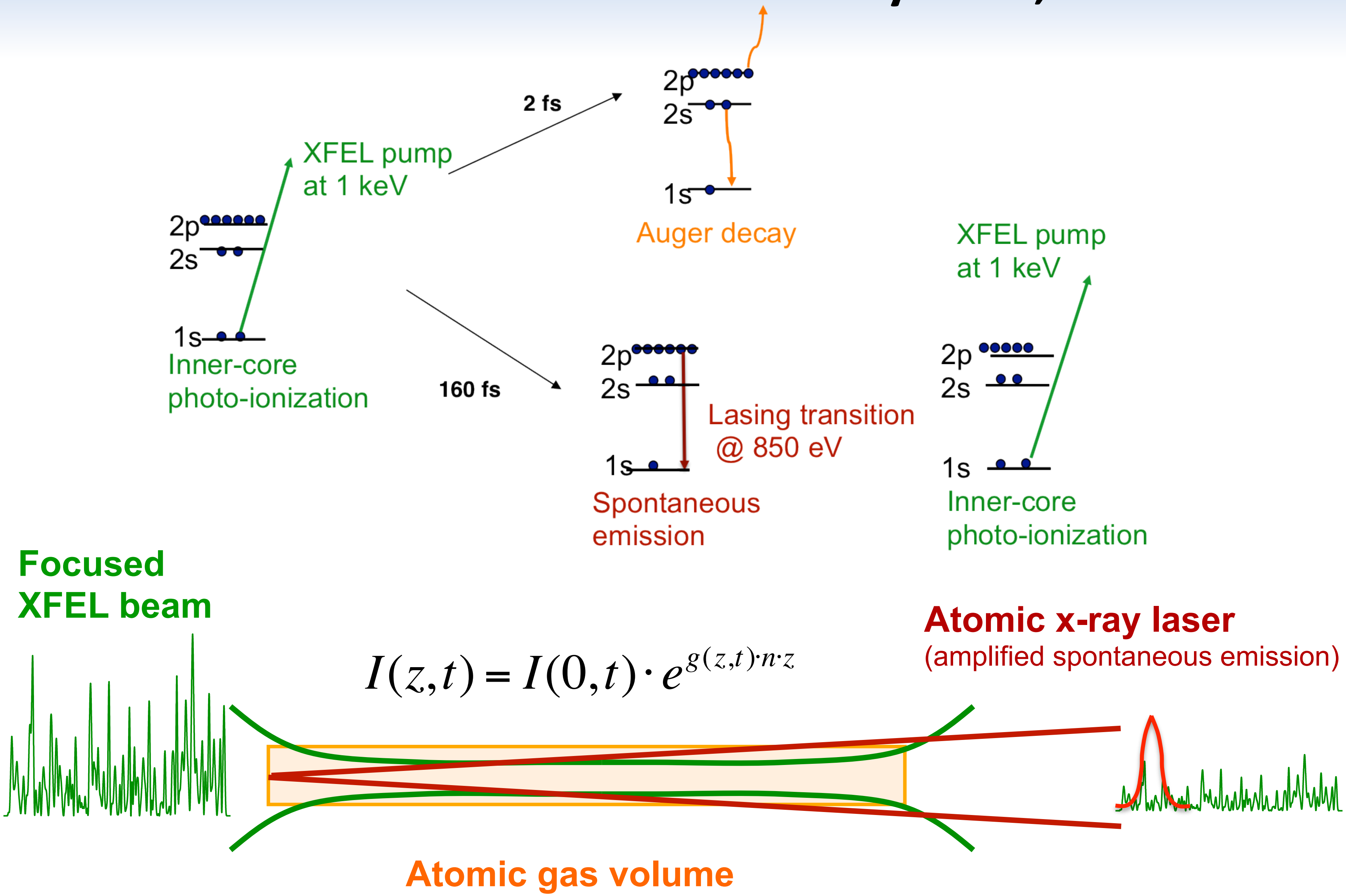
Use Synchrotrons to pump XRL

1975 (th.) Csonka and Crasemann (Li, LiH)

Use FELs to pump XRL

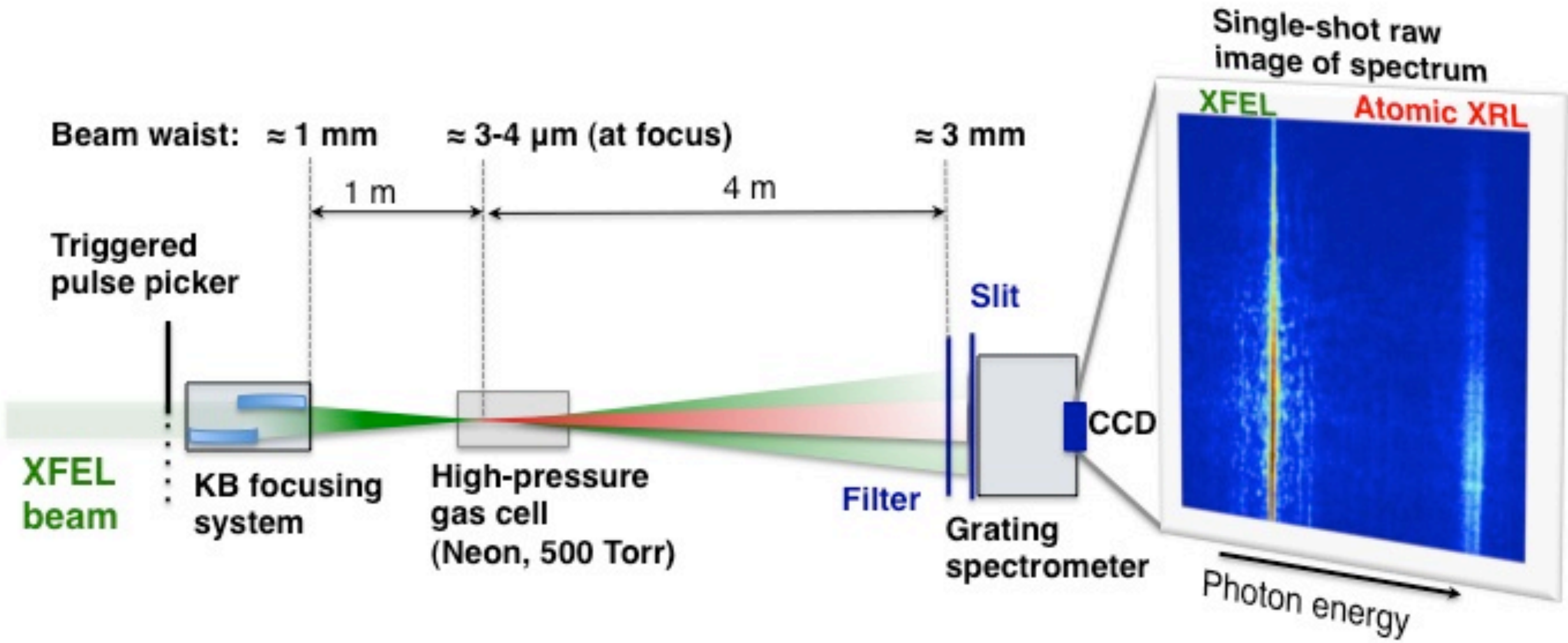
2003 (th.) Lan, Fill and Meyer-ter-Vehn (He)
2008 (th.) Zhao et al. (C, 280 eV)
2009 (th.) Rohringer and London, Phys. Rev. A 80, 013809 (Ne, 850 – 1022 eV)

Photo-ionization inner-shell x-ray laser, Neon



N. Rohringer and R. London, PRA 80, 013809 (2009)

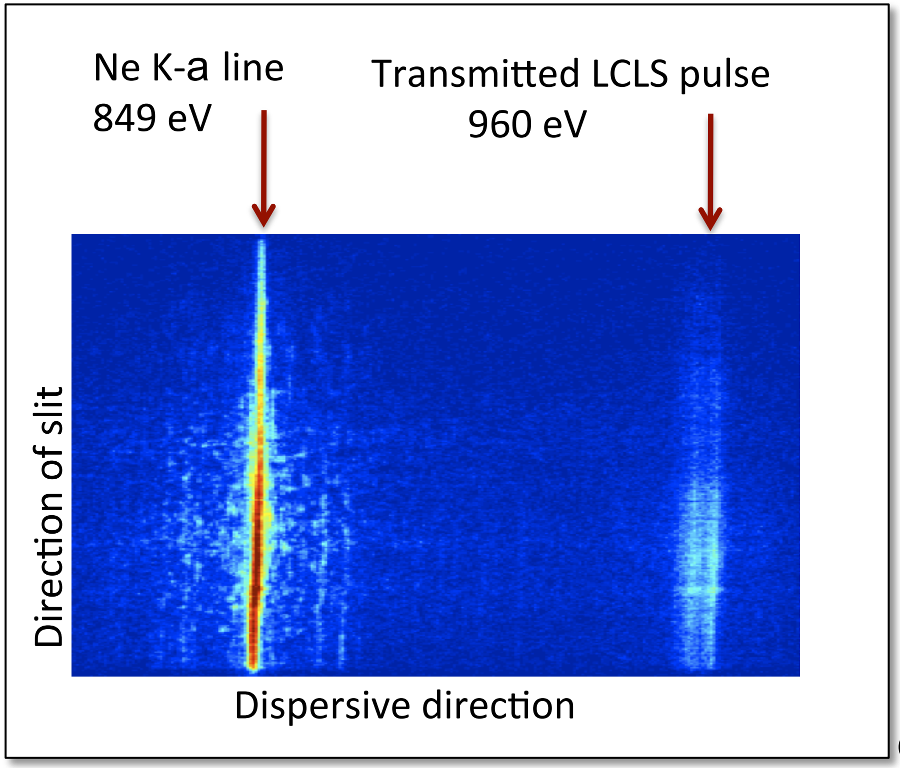
1^{st} realization of an atomic inner-shell x-ray laser
in Sept. 2010 @ LCLS



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

Single shot of highest intensity: 8×10^9 photons in Ne K-α line

corresponding to 1.1 μJ, GL 21-23



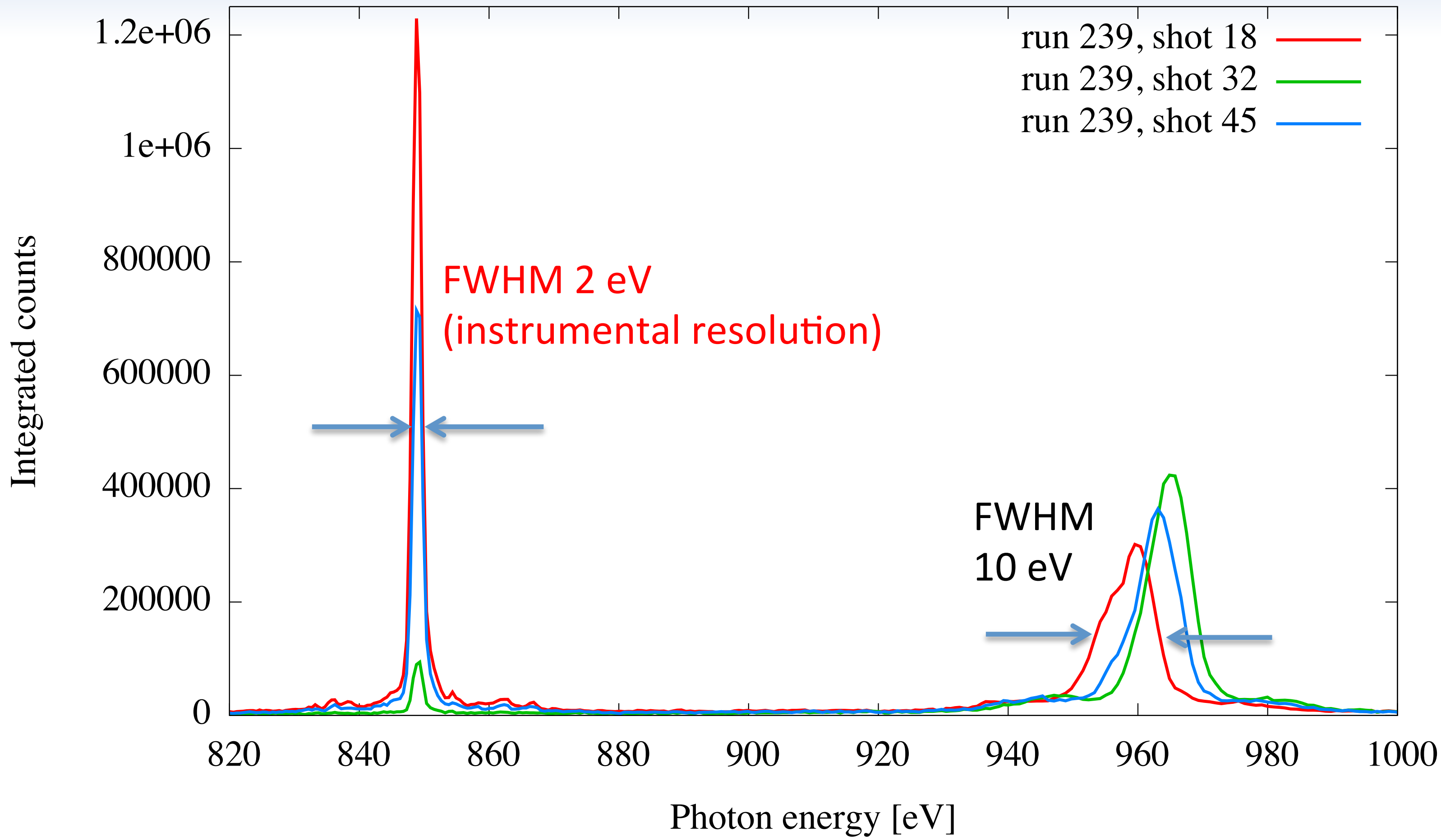
conversion efficiency:
$\approx 4 \times 10^{-3}$

Input:
LCLS pump at 960 eV
pulse energy: 1.4 mJ (0.25 mJ on target)
focus diameter: ≈ 4 micron
Pulse duration: 40 fs

Gas pressure: 500Torr
Interaction length: 1.6 cm

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

Integrated spectrum for three sample shots



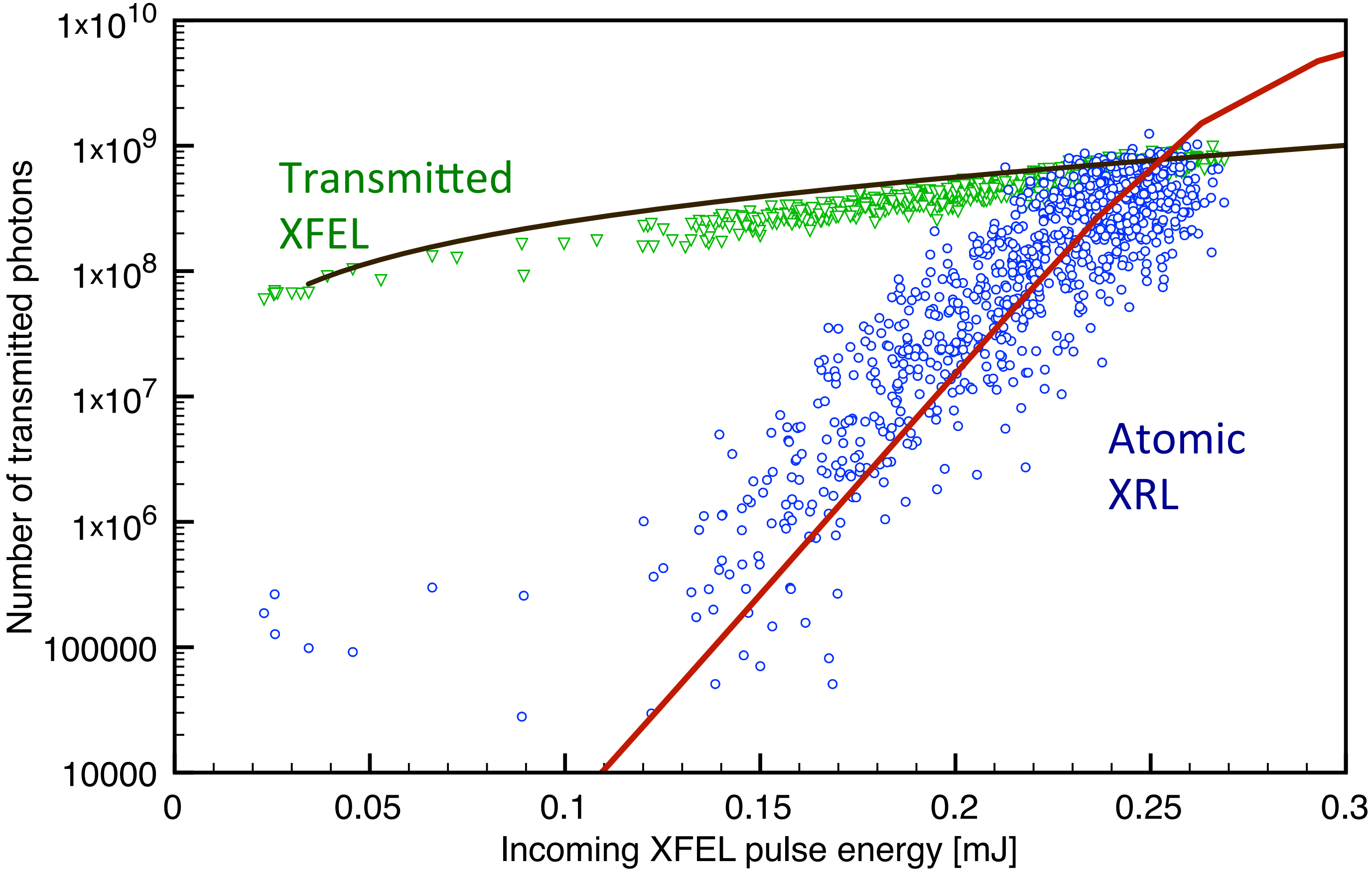
Maxwell-Bloch equations: Pulses are transform limited !!!
Pulse duration: 1-4 fs, bandwidth: 0.3 eV

Atomic inner-shell x-ray laser at 1.46 nanometres pumped by an x-ray free-electron laser, Rohringer et al., *Nature* **481**, 488 (2012)

Pumping-power dependence of Ne K-α transition

(every point corresponds to an average over 10 LCLS shots)

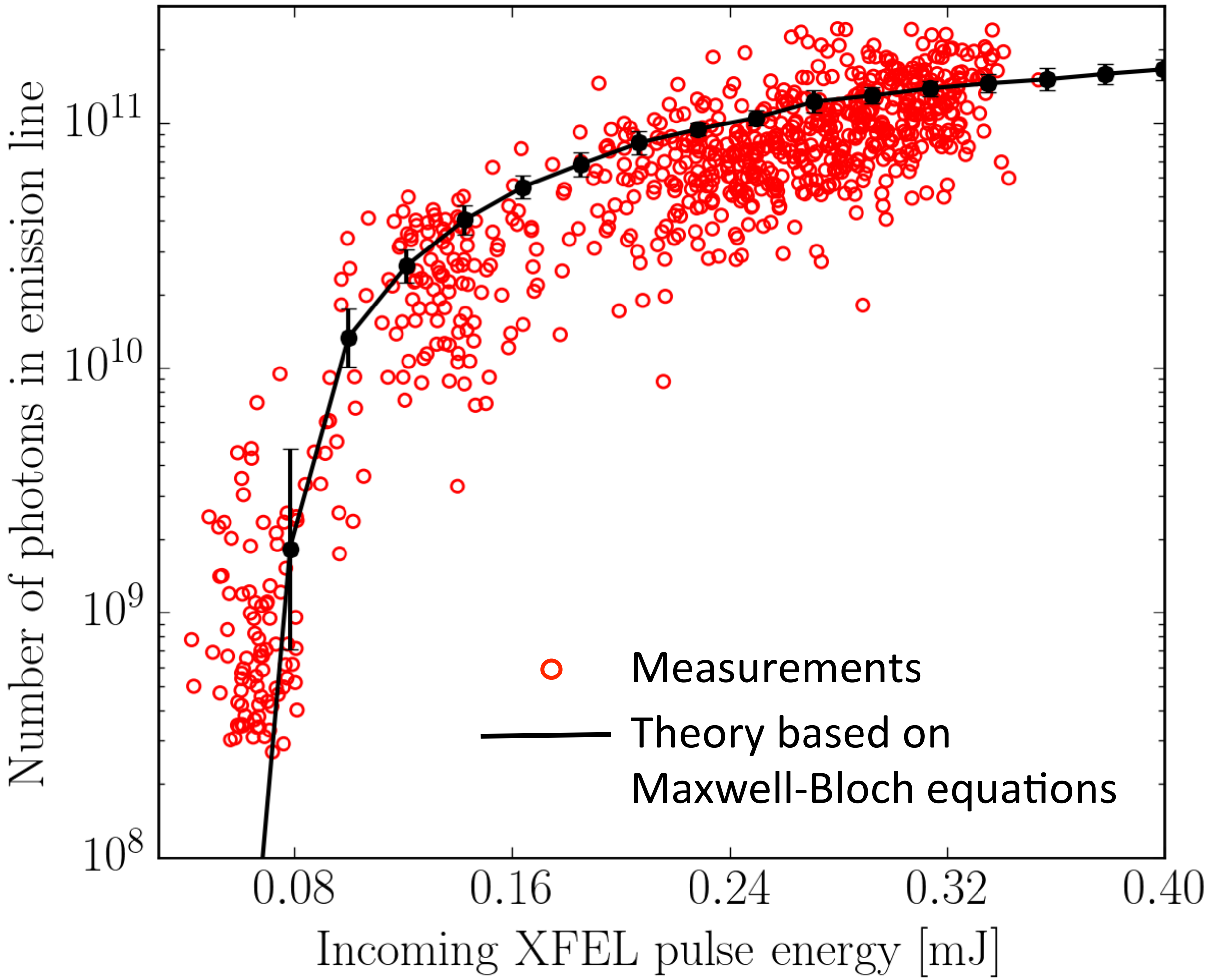
Average GL = 19-21.3 @ pulse energy of 0.25 mJ



Self-consistent gain calculations based on rate equations agree well with experiment.

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

Saturation of Ne K-α laser reached in recent experiment



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

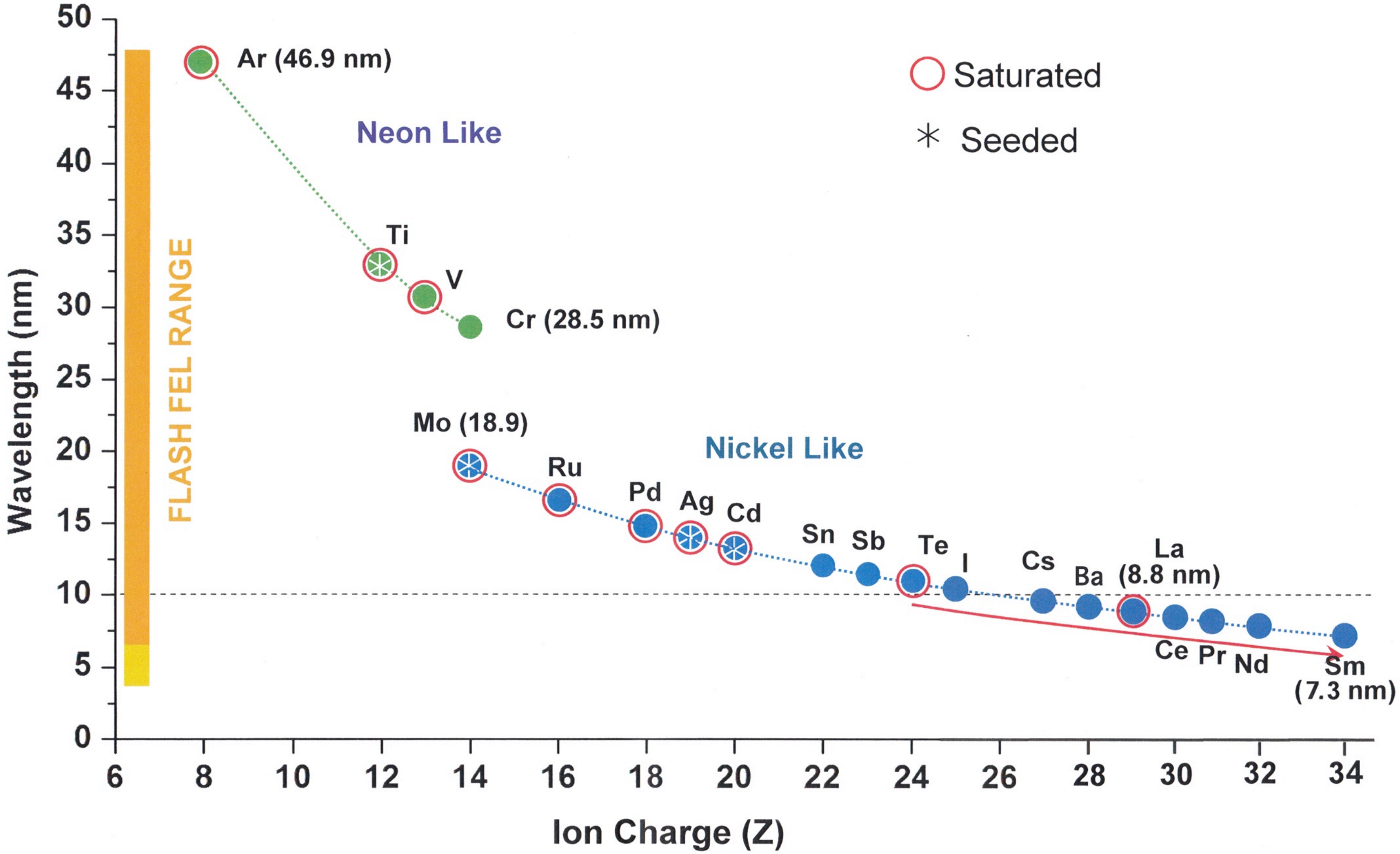
K-α x-ray lasing with other gain materials

A naïve scaling analysis

	Z	K-α Energy	Auger lifetime	1s ionization cross section	Oscillator strength	Estimated gain*
		eV	fs	cm^2		a.u.
N	7	392	7.6	6.3e-19	1.1e-1	0.8
O	8	525	5.1	4.7e-19	1.4e-1	0.7
Ne	10	849	2.7	2.9e-19	2.2e-1	0.2
S	16	2307	1.3	1.0e-19	2.9e-1	0.03

*we assumed 2 x 10^{12} photons/ 50 fs/ $(1.5\mu m)^2$ in the pump pulse

Gain-saturated table-top plasma-based soft-x-ray lasers cover 8.8 nm - 47 nm wavelength region



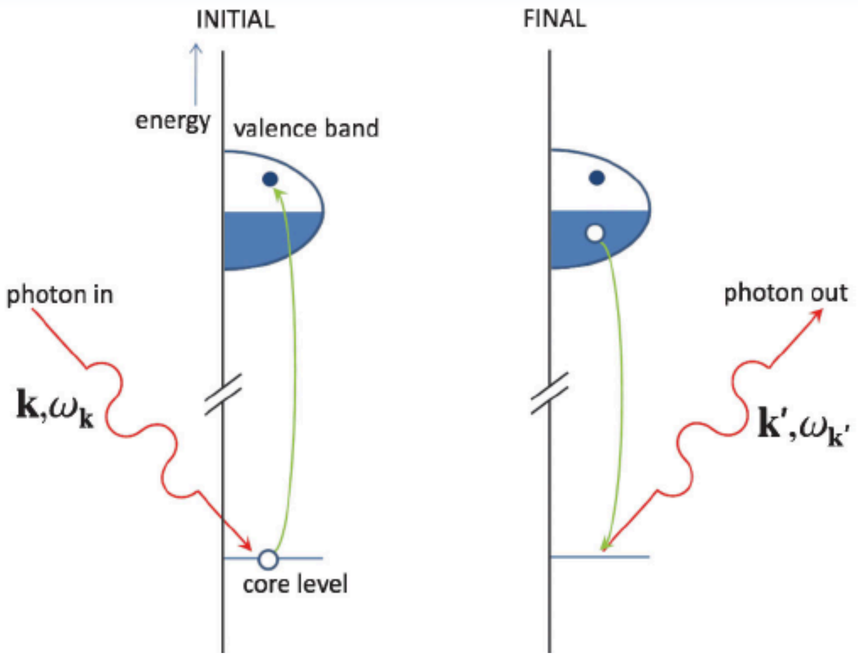
(By courtesy of J.J. Rocca, Colorado State University)

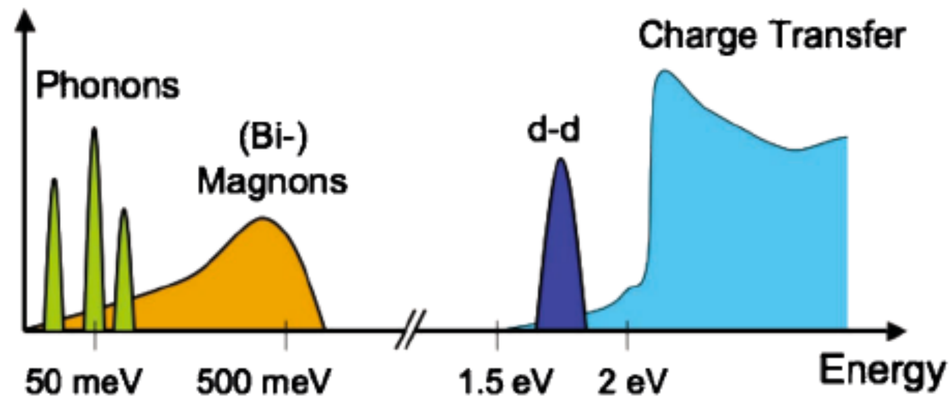
Review Articles:

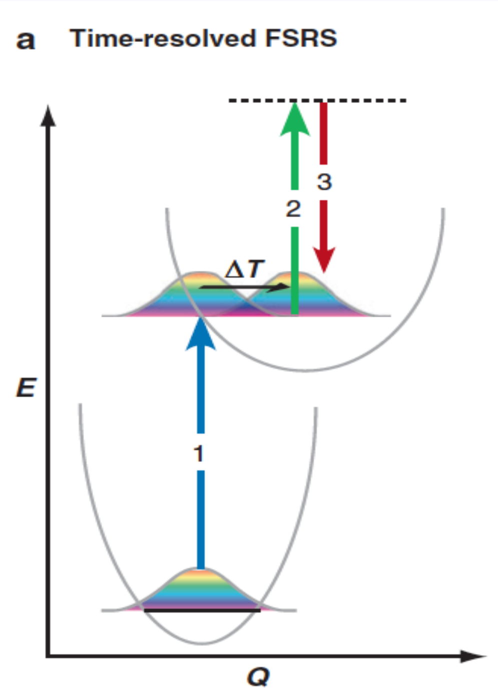
J. J. Rocca, 'Table-top soft x-ray lasers', Rev. Scient. Instr. **70**, 3799 (1999).

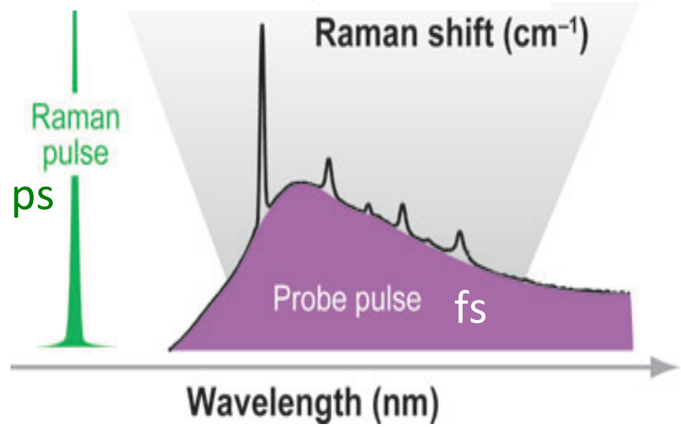
S. Suckewer and P. Jaegle, Laser Phys. Lett. **6**, No. 6, 411–436 (2009)

Nonlinear x-ray spectroscopy
Combining RIXS and time-resolved as/fs pump-probe spectroscopy





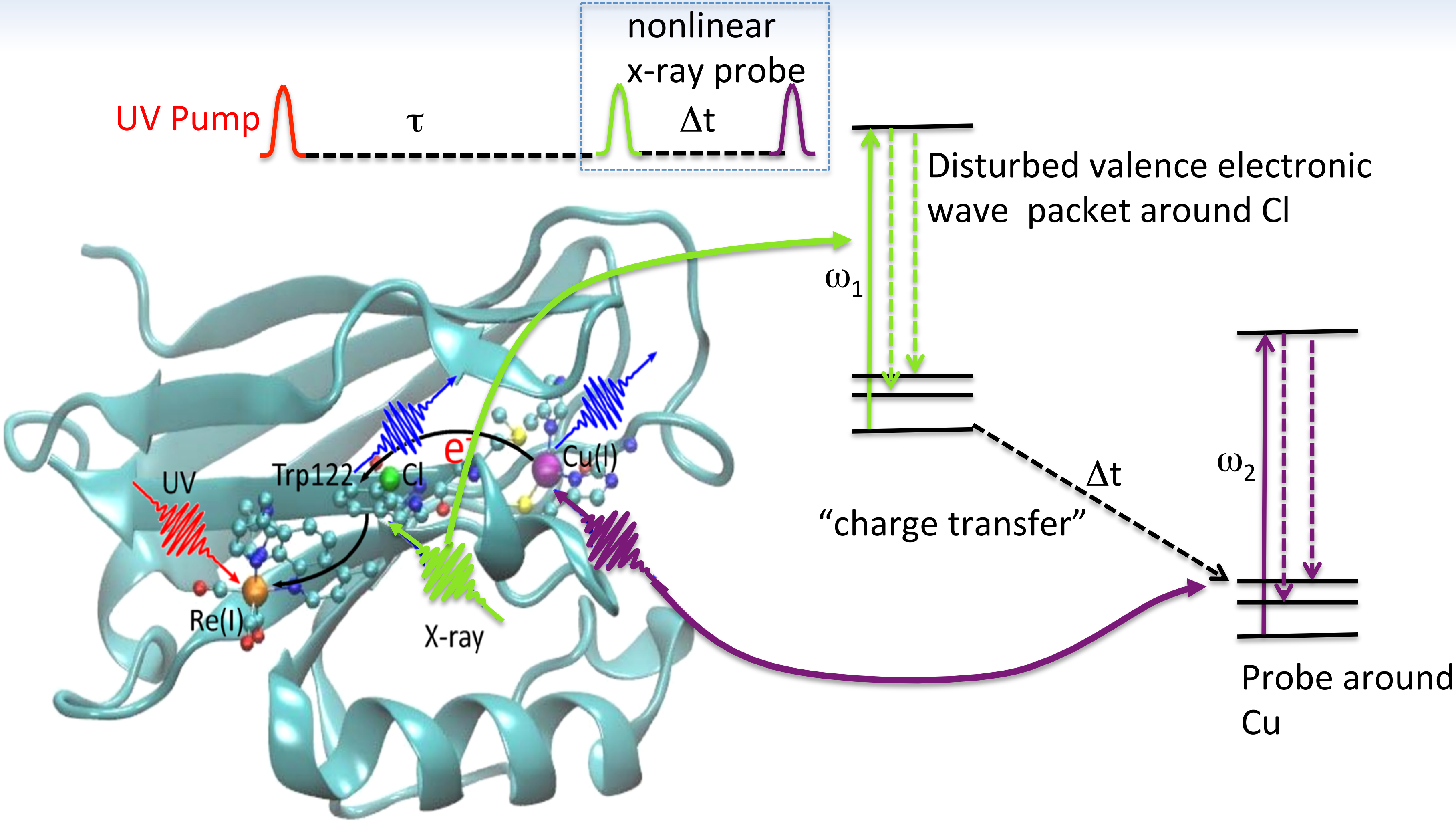




Ament et al., Rev. Mod. Phys. **83**,705 (2011)

Kukura et al., Annu. Rev. Phys. Chem. **58**, 461 (2007)

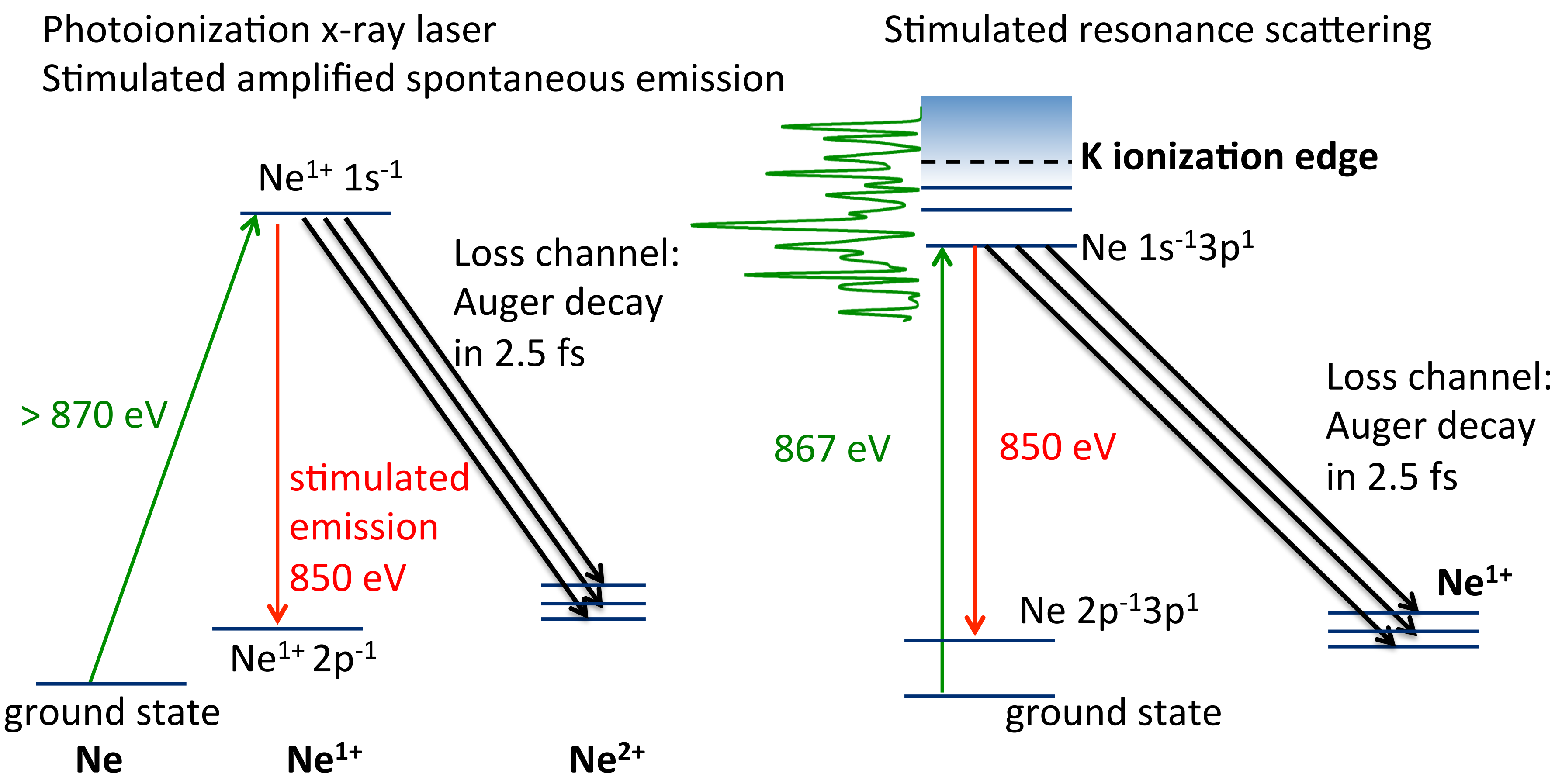
OUTLOOK: Stimulated X-ray Raman scattering a building block for nonlinear x-ray spectroscopy



Adapted from Y. Zhang et al., Phys. Chem. Lett. 5, 3656 (2014).

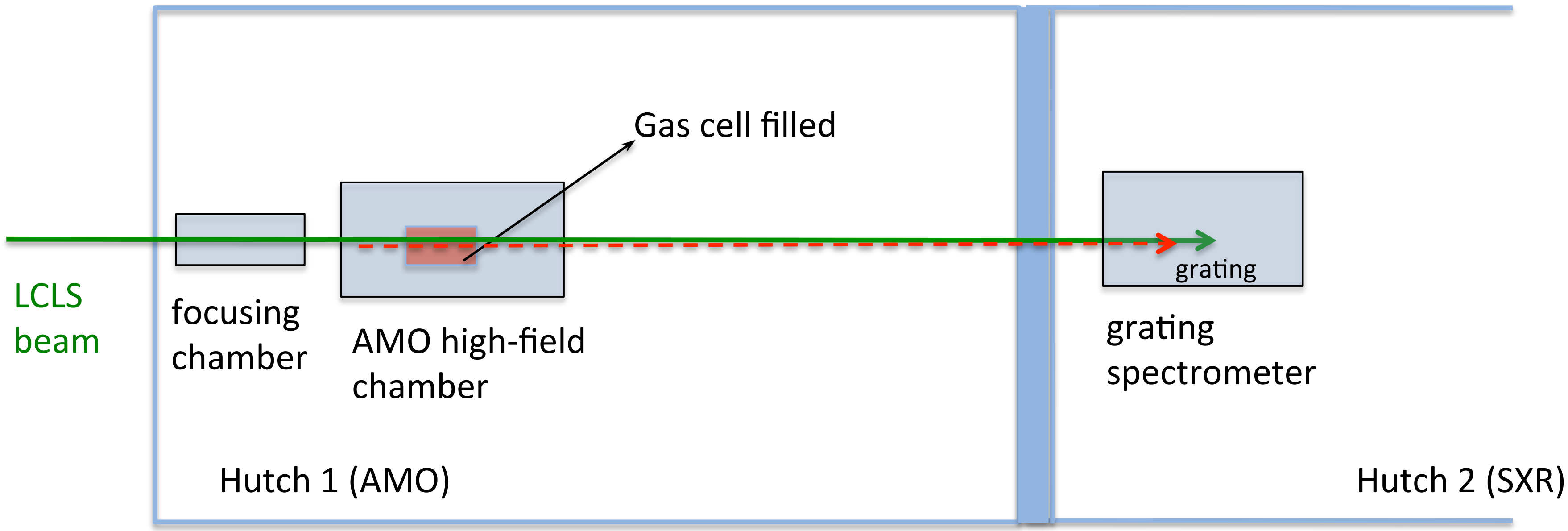
Shaul Mukamel et al. (PRL 89, 043001 (2002), PRB 72, 235110 (2005); PRA 76, 012504 (2007); PRB 79, 085108 (2009)

Stimulated electronic x-ray Raman scattering in Neon



Schematic experimental setup

2 experimental demonstrations in Aug. 2012 and Feb. 2014

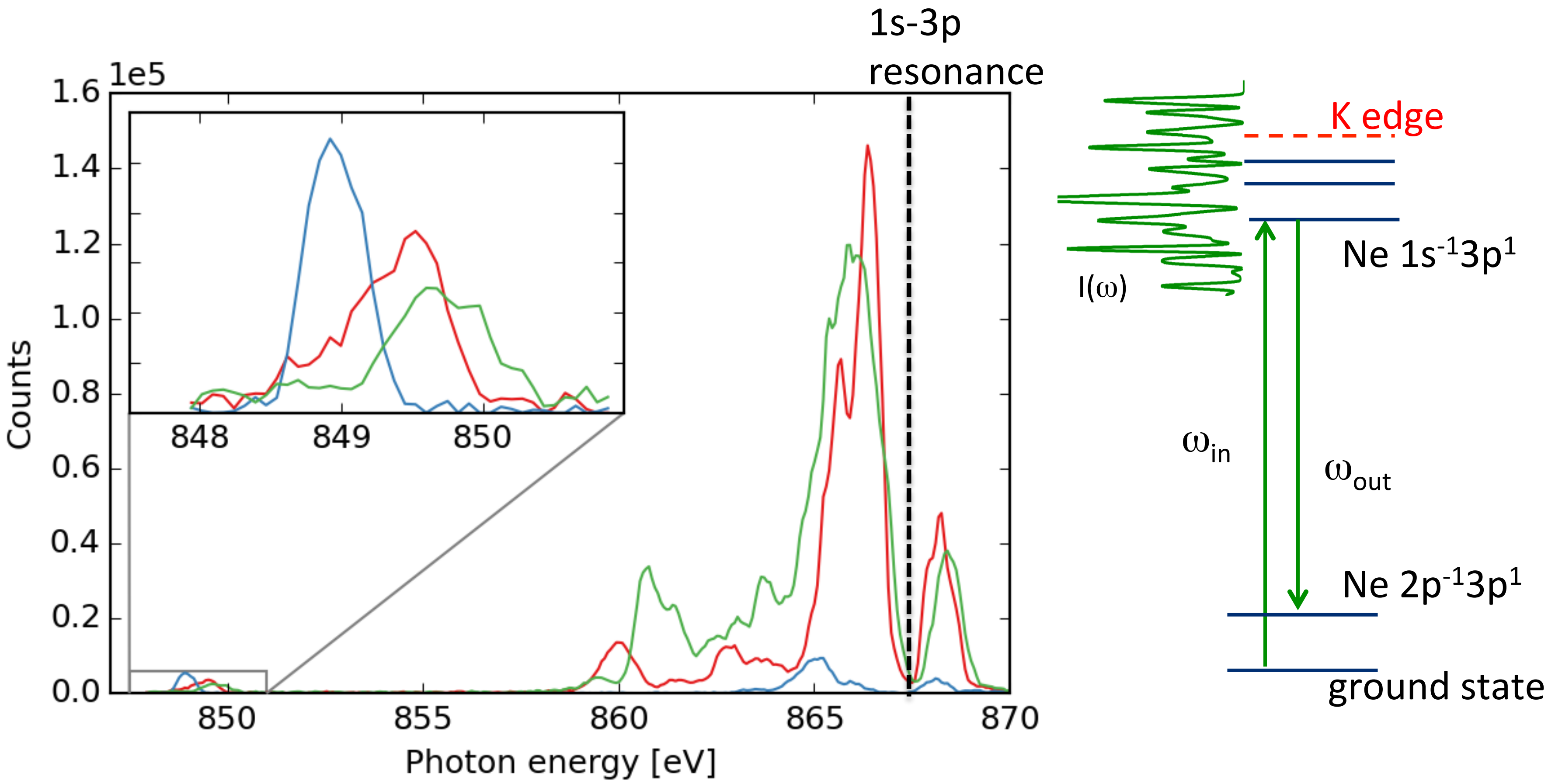


Diagnostics:

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

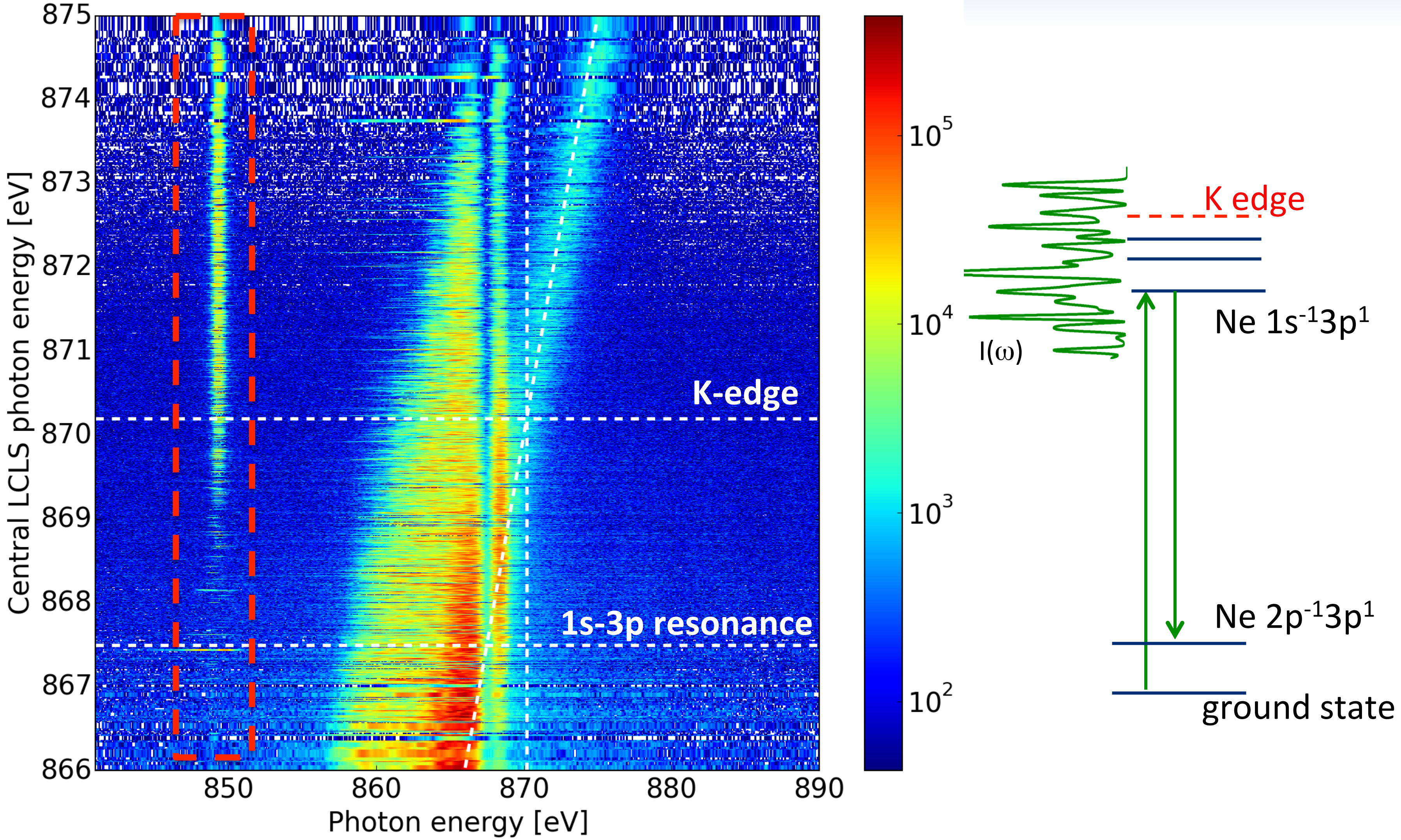
Stimulated elecronic x-ray Raman scattering in Neon (stimulated resonant inelastic x-ray scattering)

Three recorded spectra (Feb. 2014)



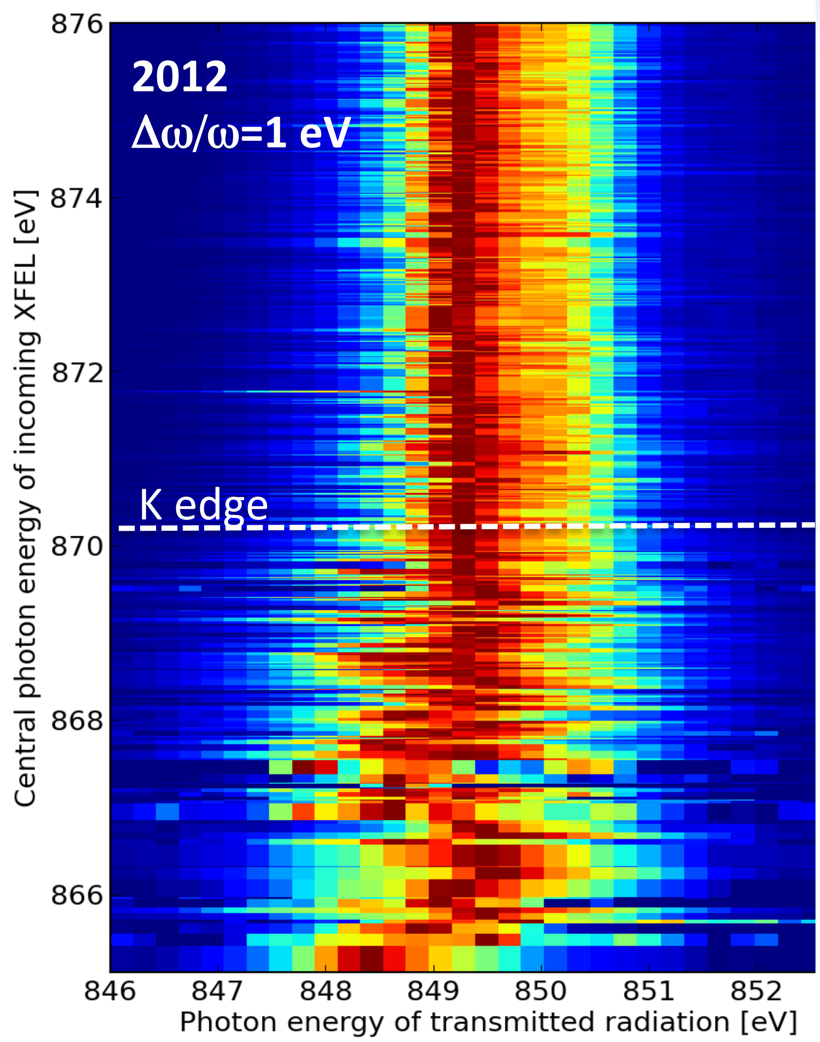
Stimulated X-Ray Raman Scattering with Free-Electron Laser Sources, N. Rohringer et al., submitted (2015).
Stimulated Electronic X-Ray Raman Scattering, C. Weninger, N.R. et al., Phys. Rev. Lett. **111**, 233902 (2013)

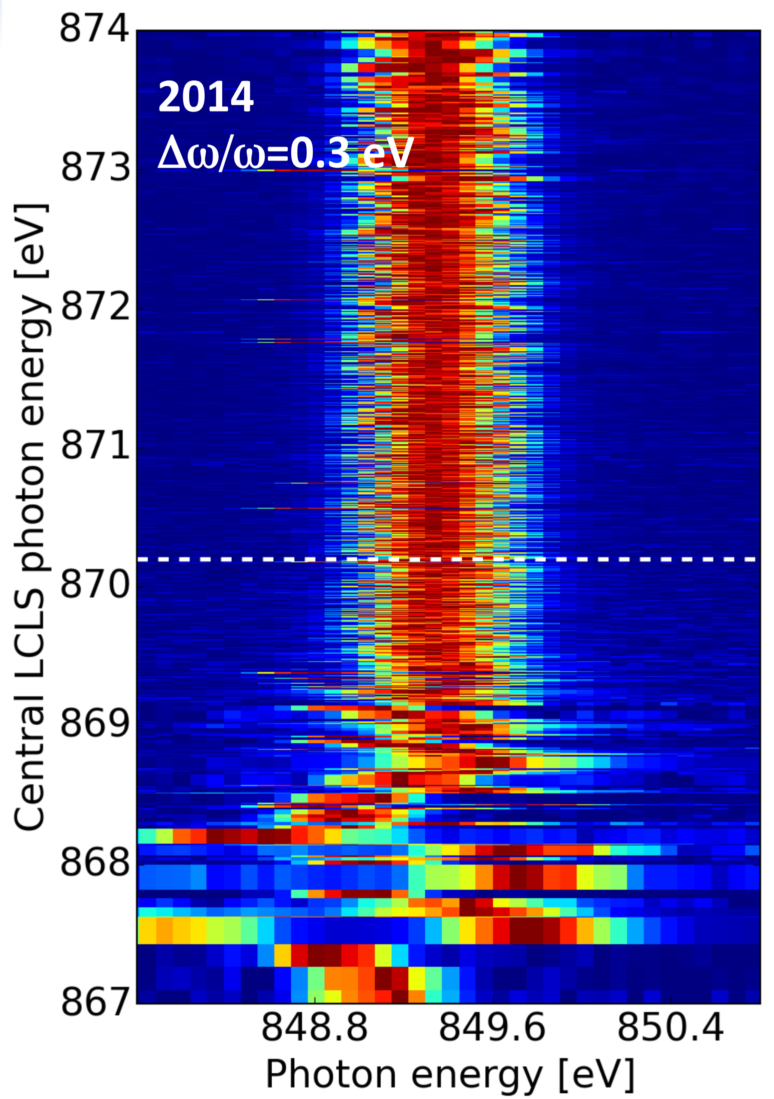
Stimulated elecronic x-ray Raman scattering in Neon (stimulated resonant inelastic x-ray scattering)



Stimulated X-Ray Raman Scattering with Free-Electron Laser Sources, N. Rohringer et al., submitted (2015).
Stimulated Electronic X-Ray Raman Scattering, C. Weninger, N.R. et al., Phys. Rev. Lett. **111**, 233902 (2013)

Emitted line profile as a function of pump photon energy

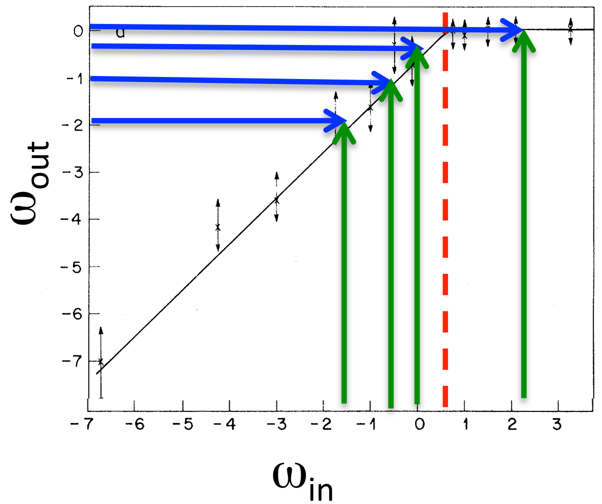




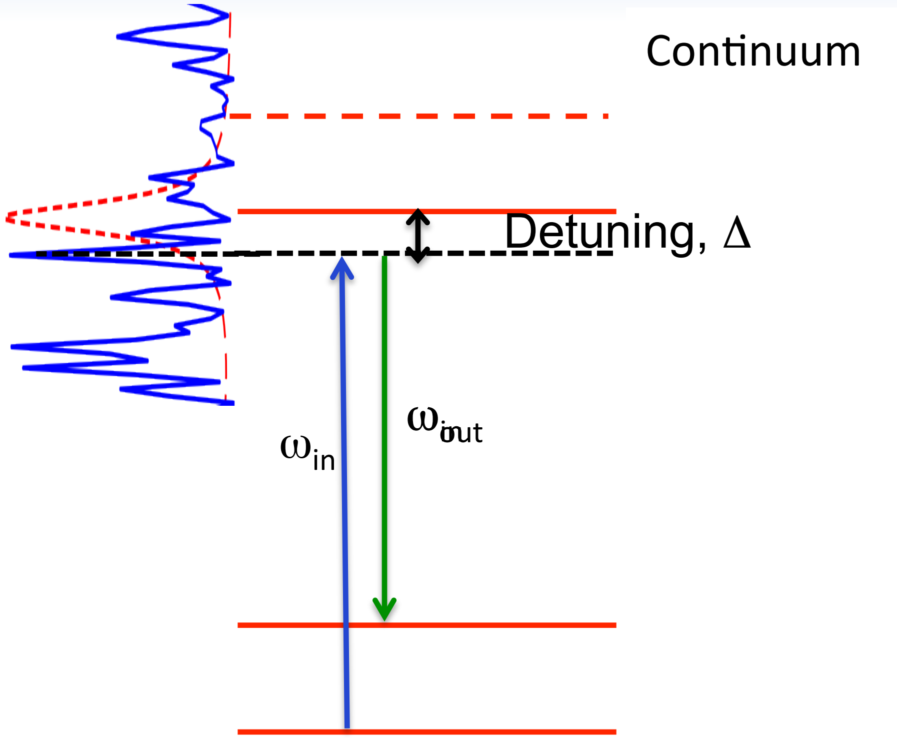
Stimulated X-Ray Raman Scattering with Free-Electron Laser Sources, N. Rohringer et al., submitted (2015).
Stimulated Electronic X-Ray Raman Scattering, C. Weninger, N.R. et al., Phys. Rev. Lett. **111**, 233902 (2013)

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

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



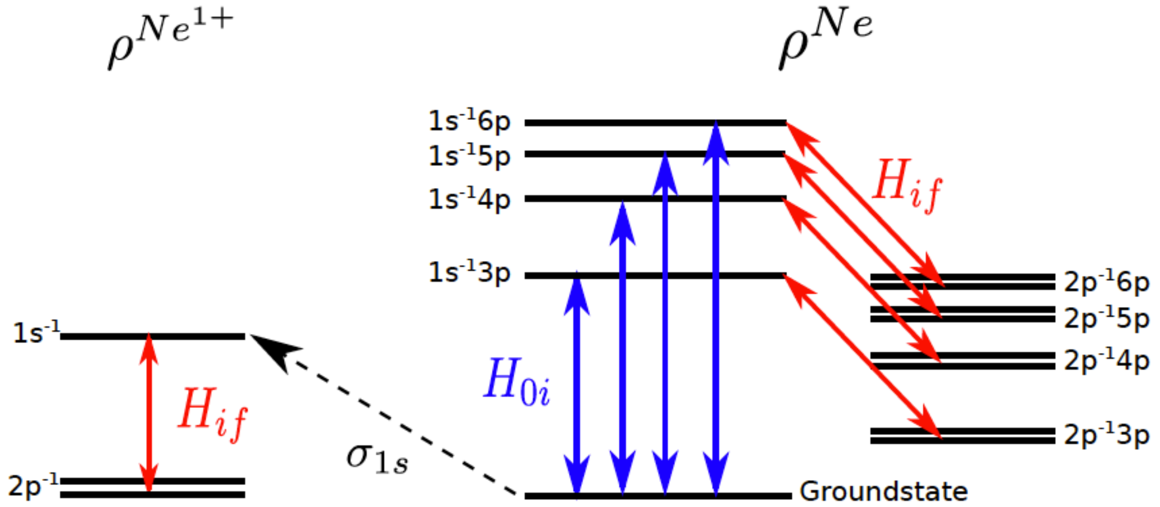
P. Eisenberger, P.M. Platzman, H. Winick, PRL 36, 623 (1976).



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



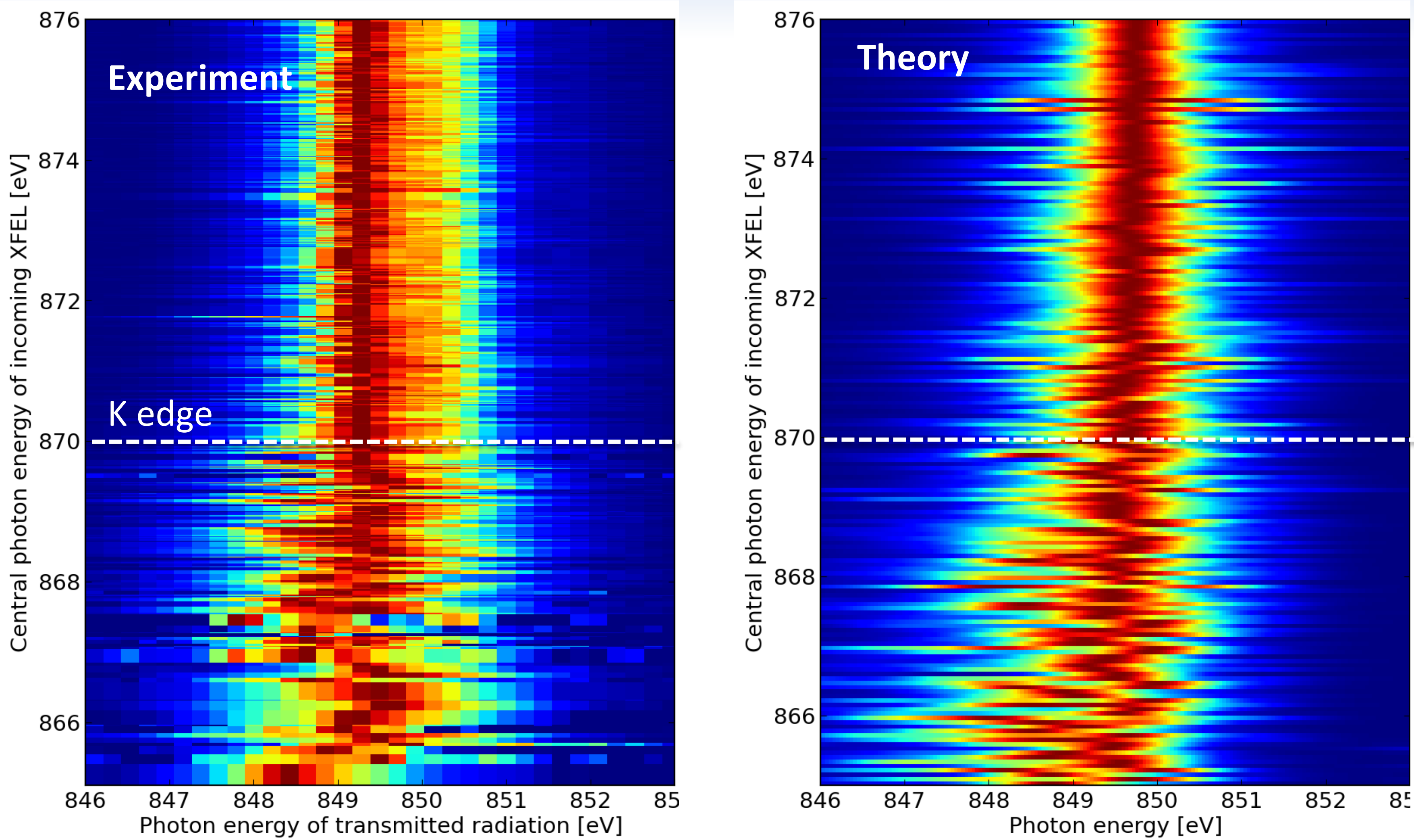
$$\frac{\partial}{\partial t}\hat{\rho} = -i[\hat{H},\hat{\rho}] - \hat{\Gamma}\hat{\rho} - \sigma\hat{\rho}I_p + S$$

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

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

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

Line profile – comparison of experiment to simulation



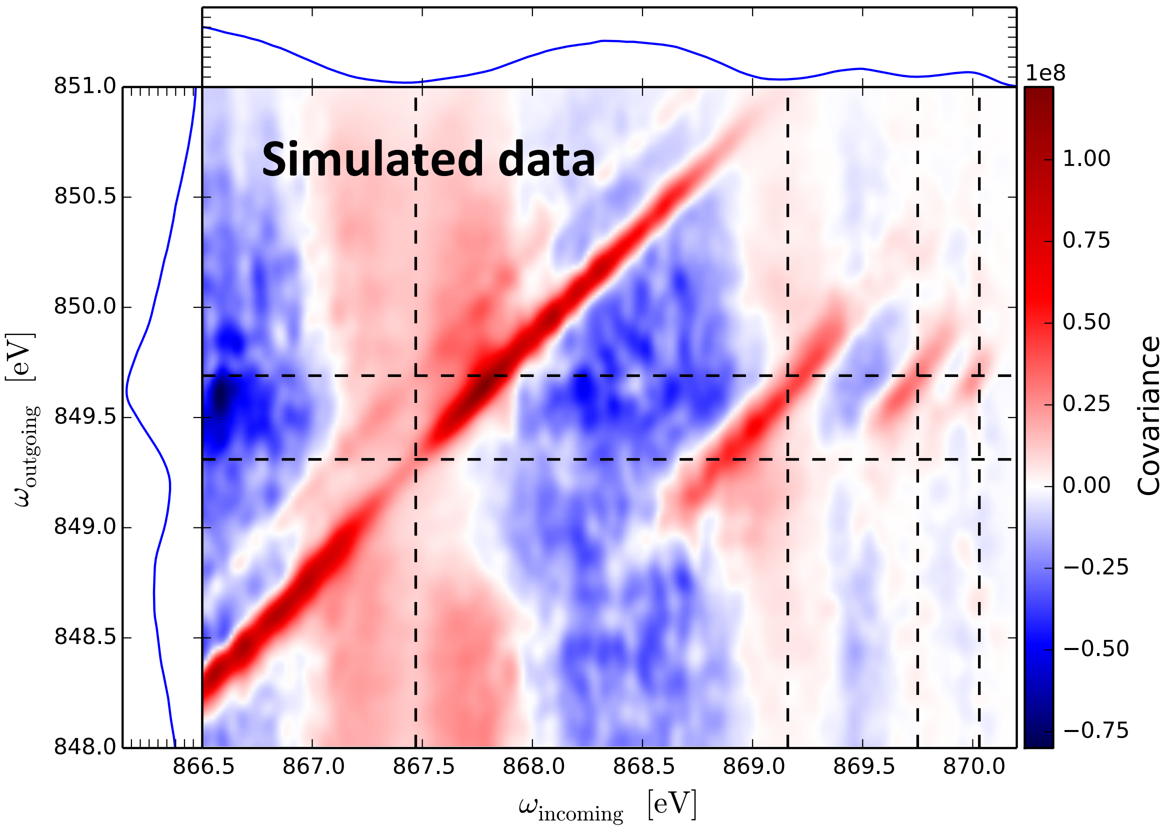
Stimulated Electronic X-Ray Raman Scattering,
C. Weninger, N.R. et al., Phys. Rev. Lett. **111**, 233902 (2013)

Stimulated resonant x-ray Raman scattering with incoherent radiation,
C. Weninger & N.R., Phys. Rev. A 88, 053421 (2013).

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

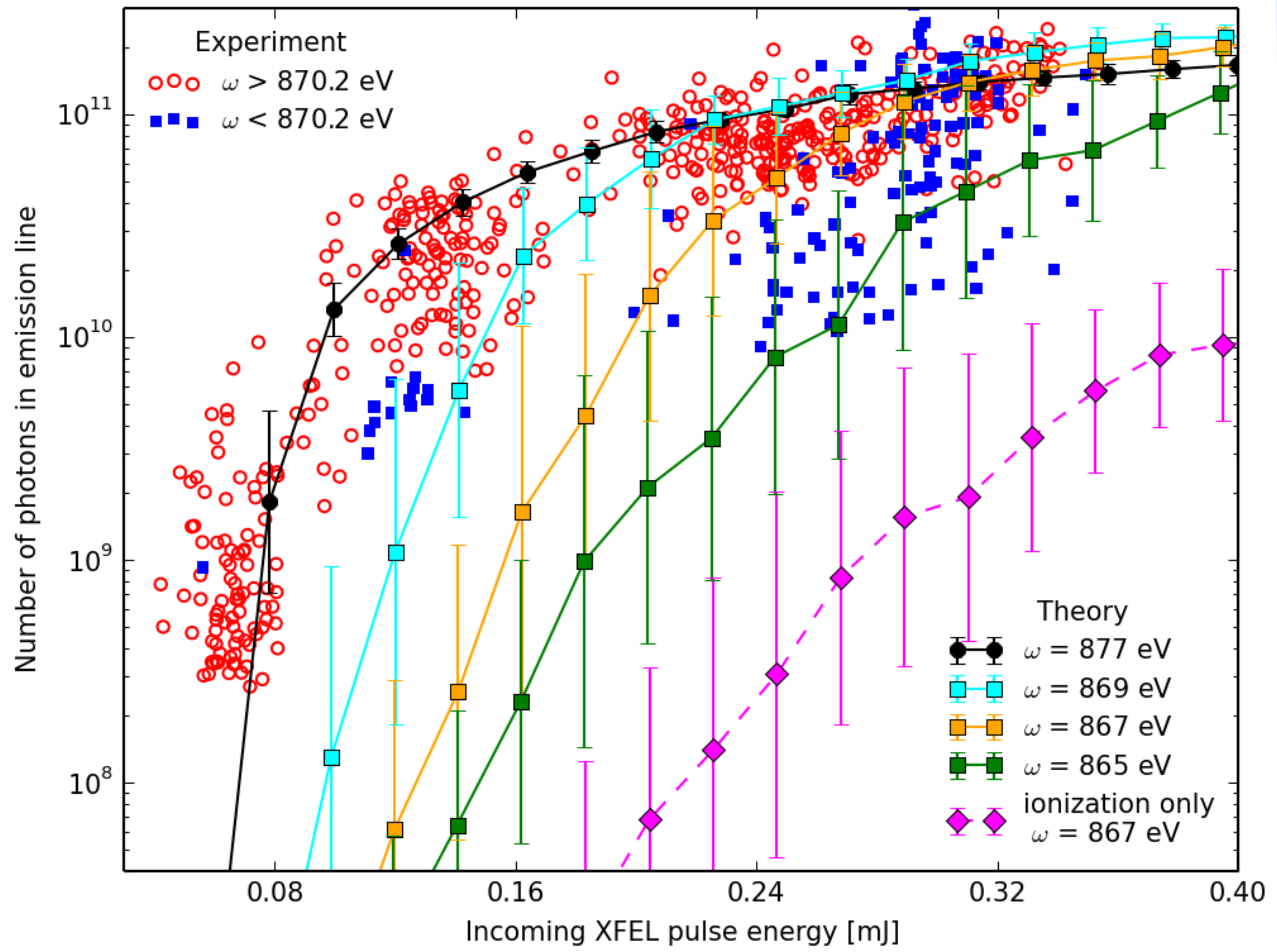
$$Cov(\omega_1, \omega_2) = <I(\omega_1)I(\omega_2)> - <I(\omega_1)><I(\omega_2)>$$

Covariance map from 5000 simulated single-shot spectra



Stimulated resonant x-ray Raman scattering with incoherent radiation, C. Weninger & N.R., Phys. Rev. A 88, 053421 (2013).

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)

Summary and Outlook

Use full potential of XFELs to exploit **quantum optics** and **nonlinear processes** in the x-ray spectral domain:

Coherently amplify x-ray emission to boost signal for measuring high resolution spectra in a single shot

- x-ray emission spectroscopy
- resonant inelastic x-ray scattering

Explore possibility of nonlinear x-ray spectroscopy to follow electron dynamics in a coherent way

-> coherent nonlinear x-ray spectroscopic techniques

  - attosecond coherent x-ray Raman spectroscopy
  - x-ray four-wave mixing
  - transient grating spectroscopies
  - Auger-electron spectroscopy as "read-out" of nonlinear susceptibility