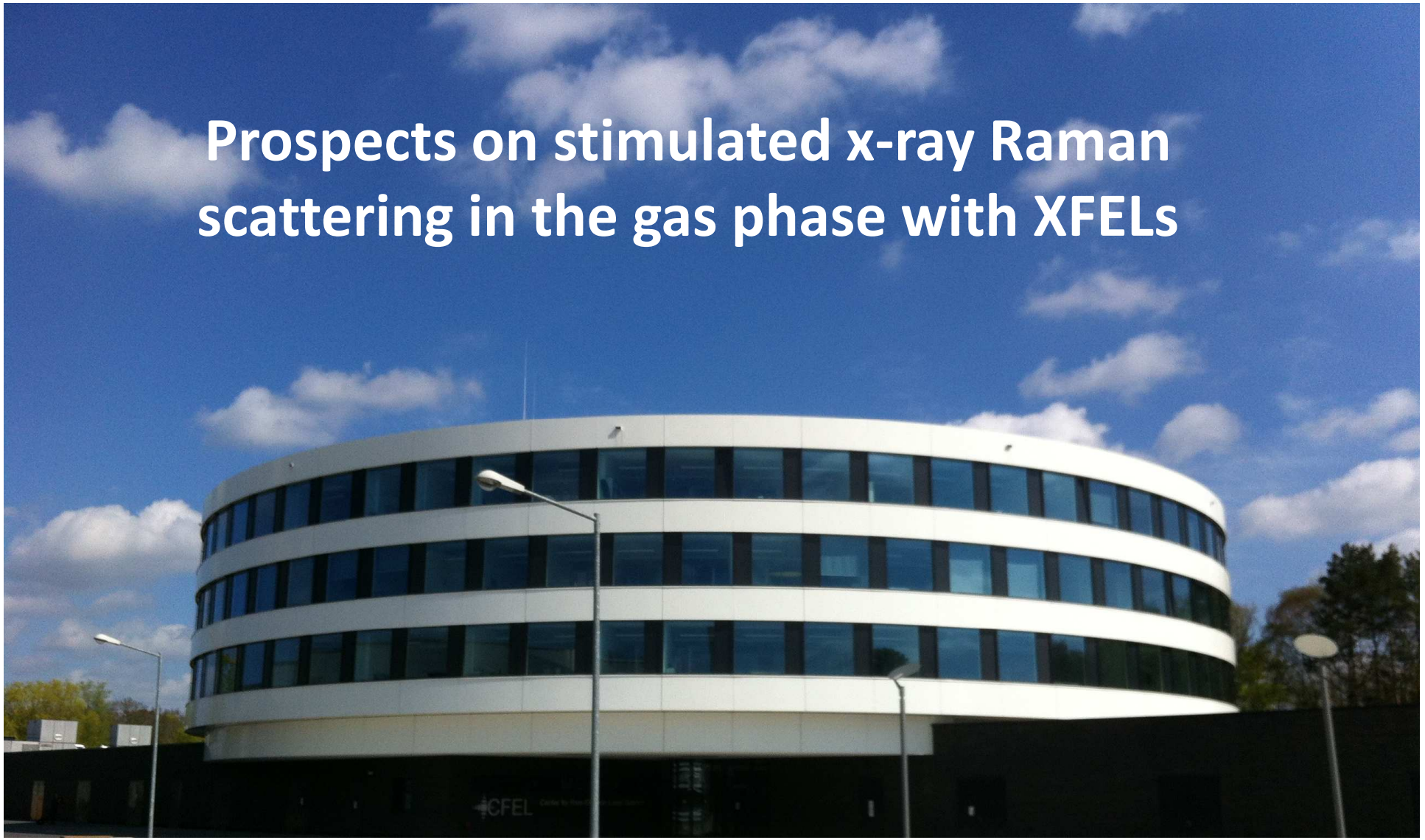
Prospects on stimulated x-ray Raman scattering in the gas phase with XFELs



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Center for Free-Electron Laser Science, Hamburg

CFEL SCIENCE

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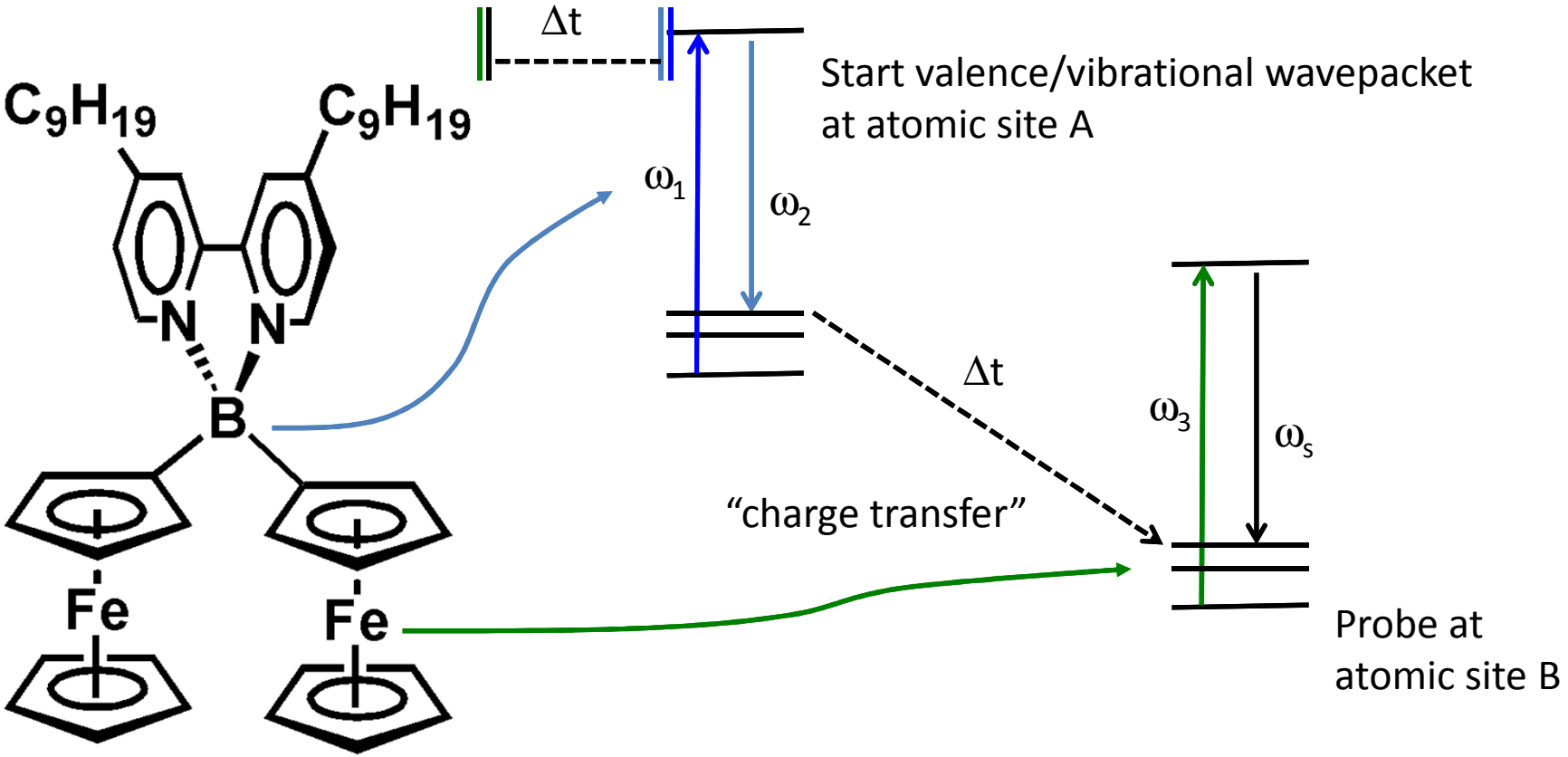
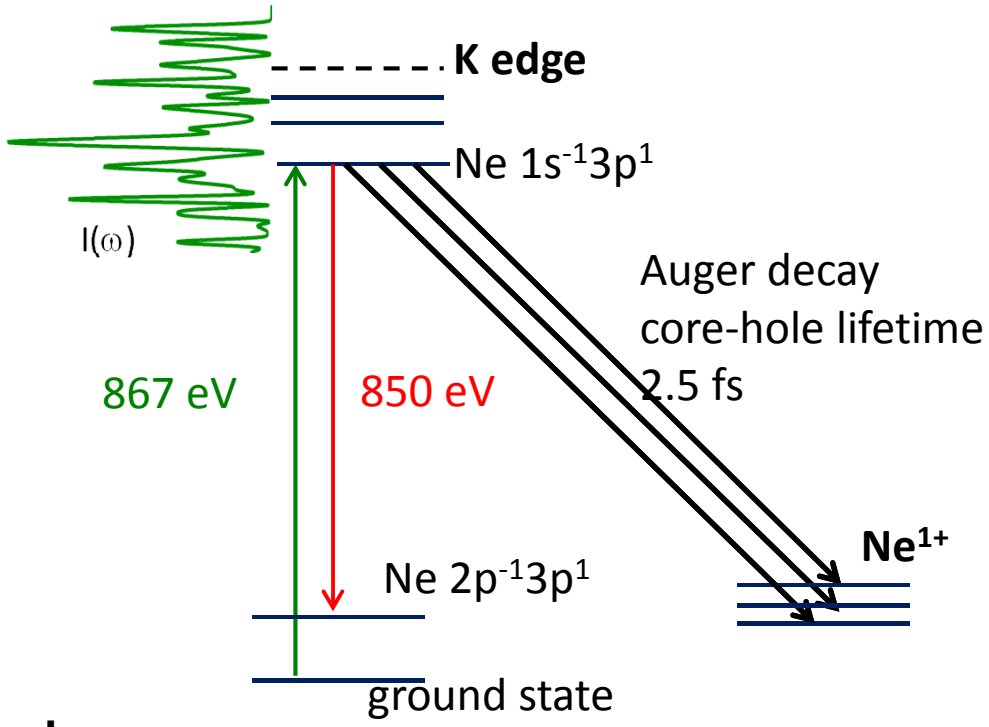
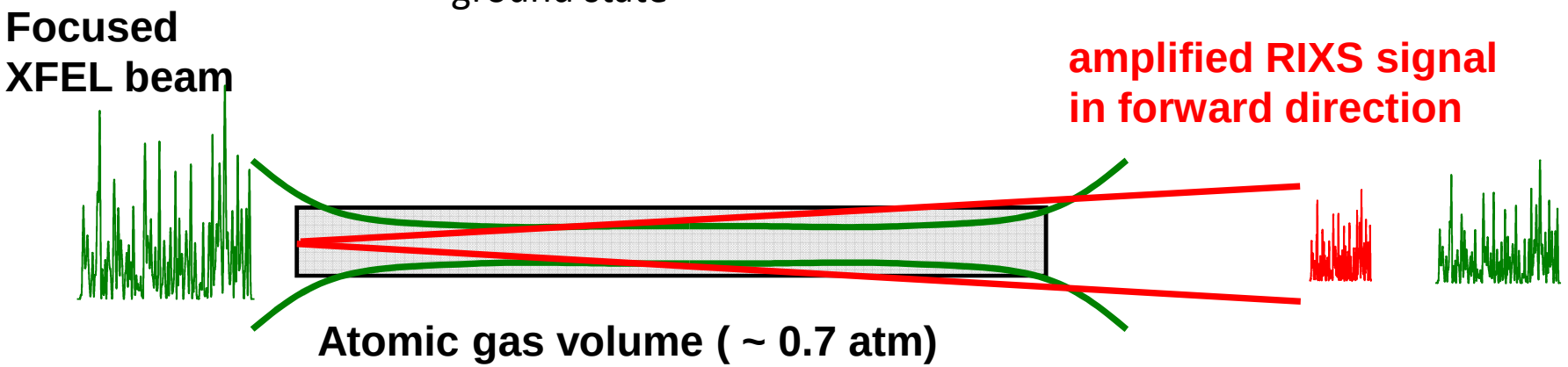
Stimulated X-ray Raman scattering
a building block for nonlinear x-ray spectroscopy



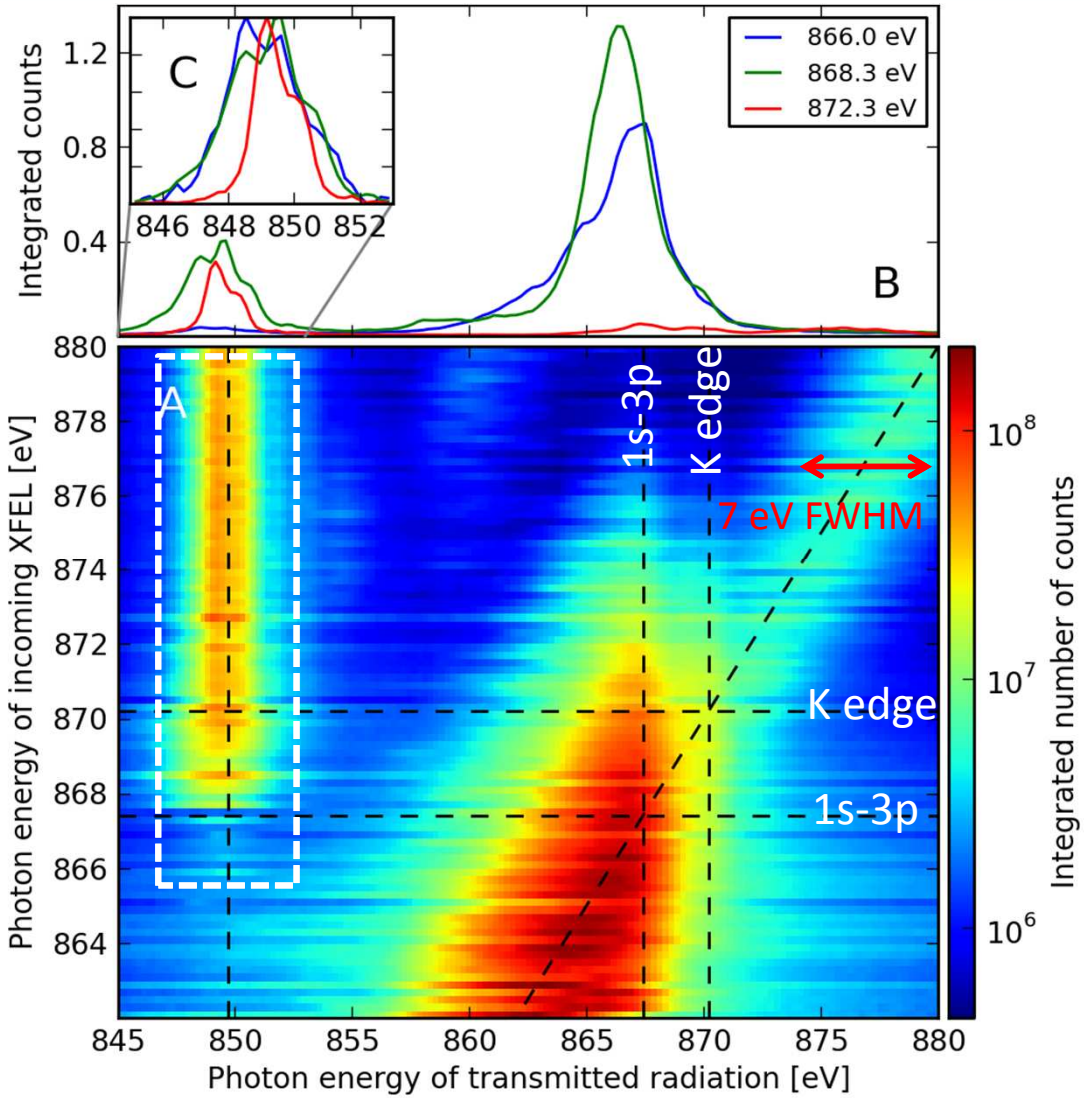
S. Mukamel et al. (PRL 89, 043001 (2002), PRB 72, 235110 (2005); PRA 76, 012504 (2007); PRB 79, 085108 (2009)

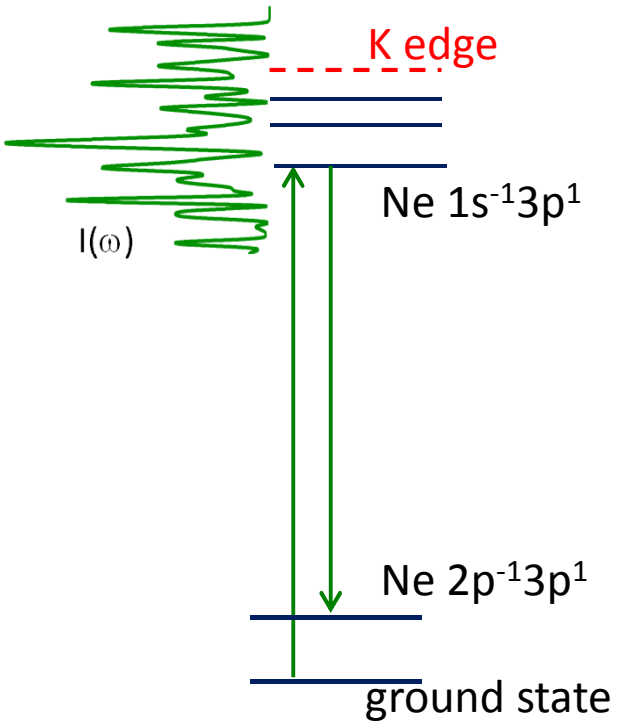
Stimulated resonant inelastic x-ray scattering in optically dense gas samples





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



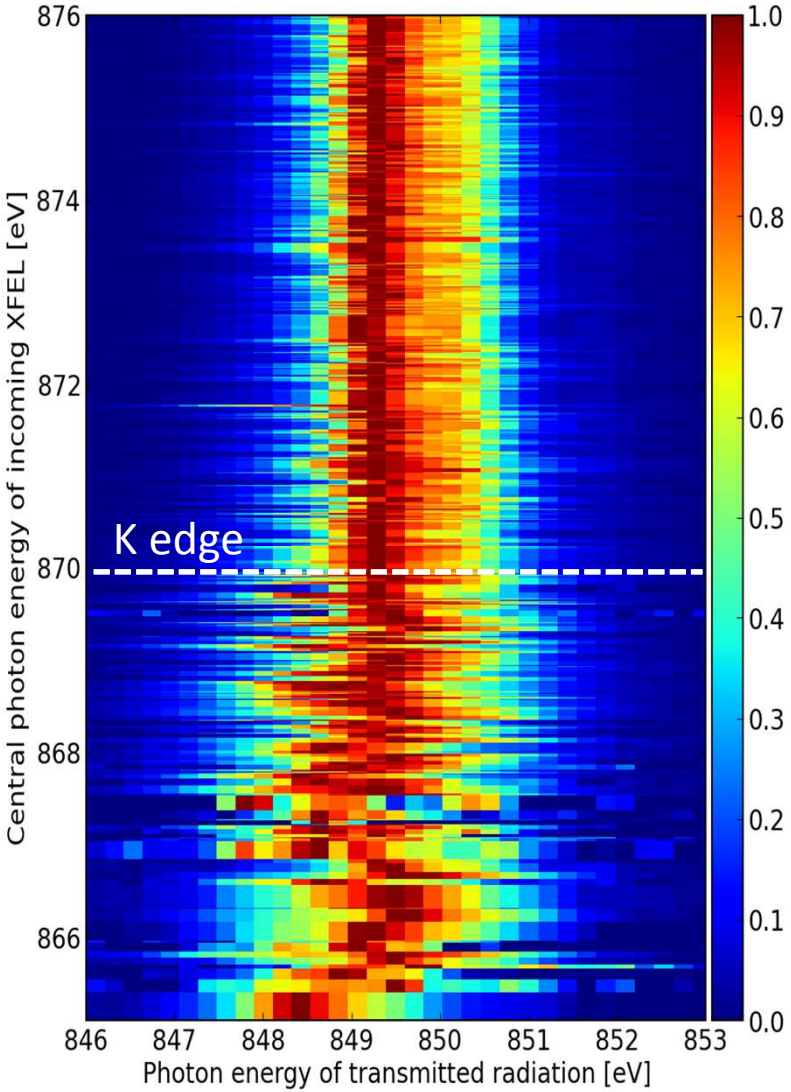


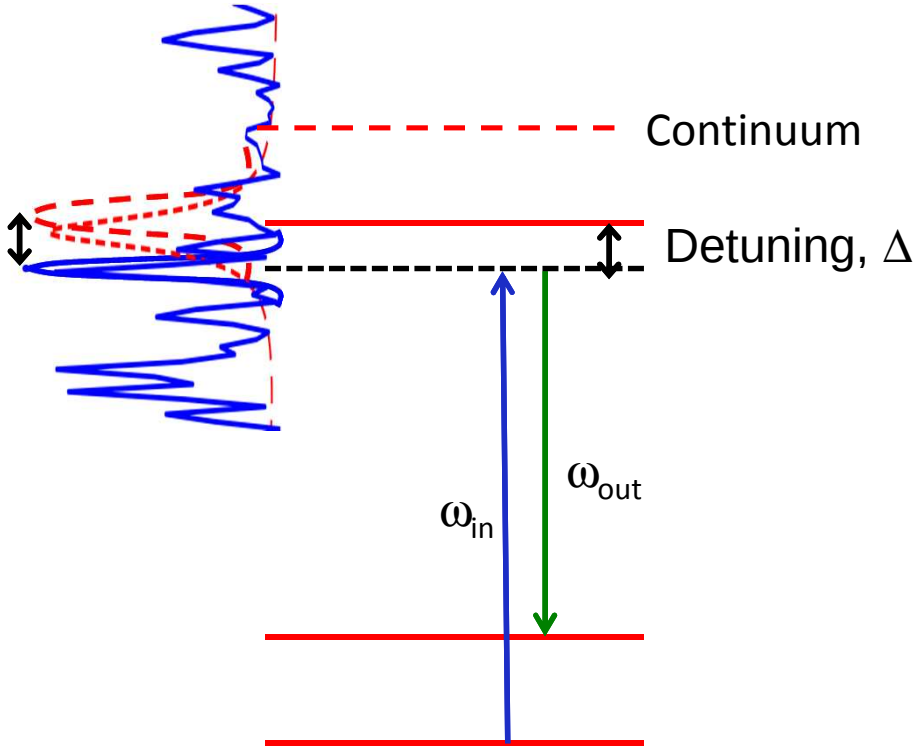
C. Weninger et al.,
Phys. Rev. Lett. **111**, 233902 (2013)

Emitted line profile as a function of pump photon energy

Spiky SASE spectrum creates stochastic shifts

("anomalous" linear dispersion of resonance scattering)





Width of resonance: 0.25 eV

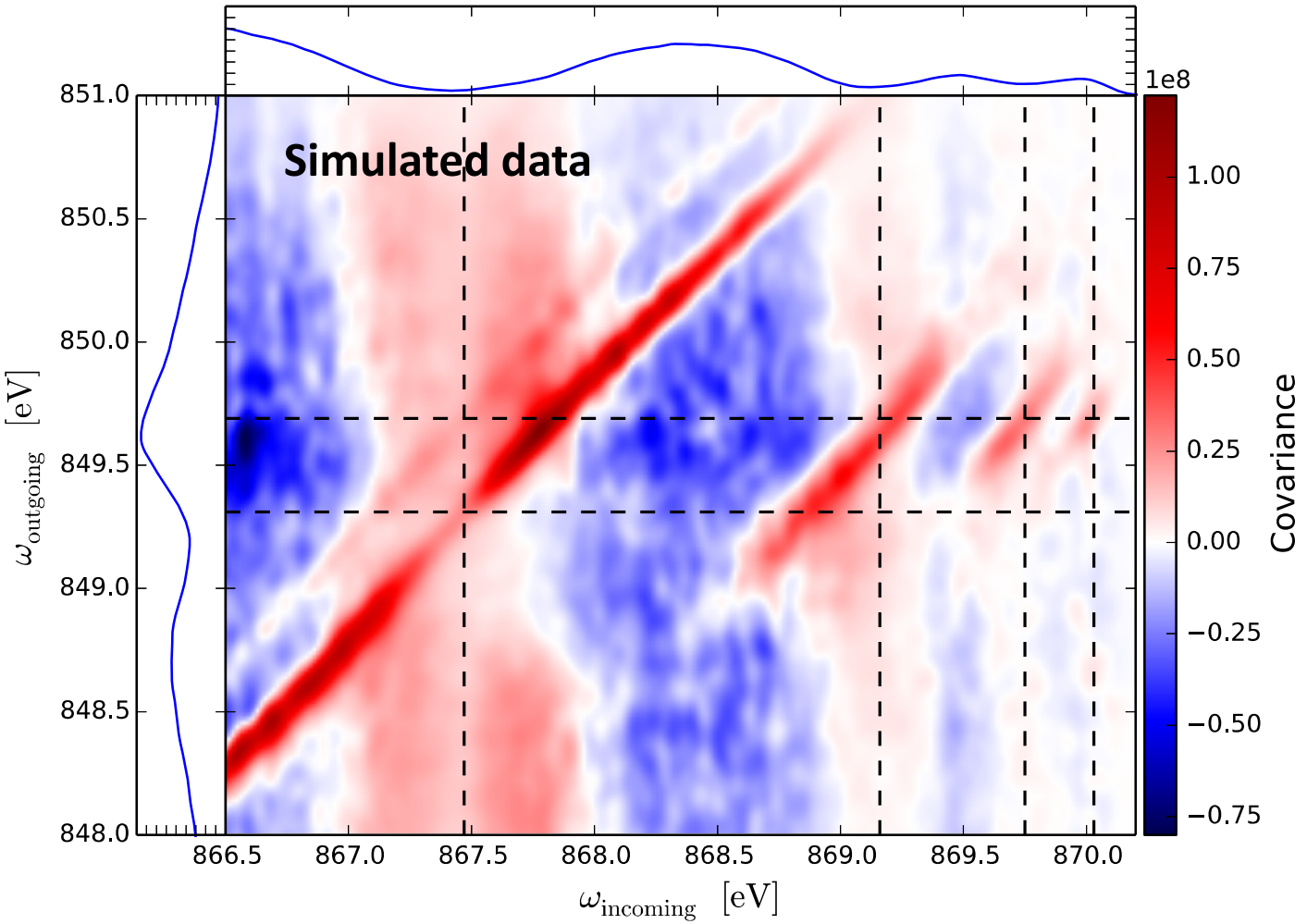
Width of SASE spike: $\Delta\omega = 1/\tau = 0.1$ eV

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

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

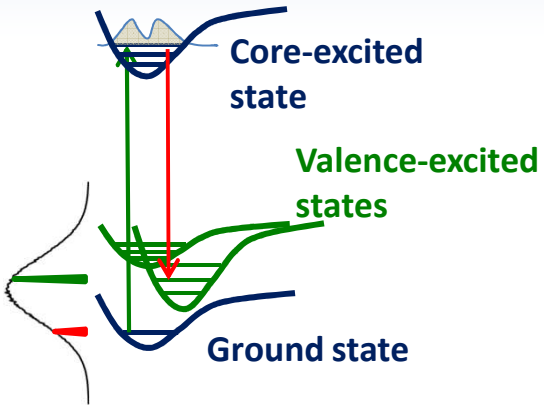
$$Cov(\omega_1, \omega_2) = < I(\omega_1) I(\omega_2) > - < I(\omega_1) >< I(\omega_2) >$$

Covariance map from 5000 simulated single-shot

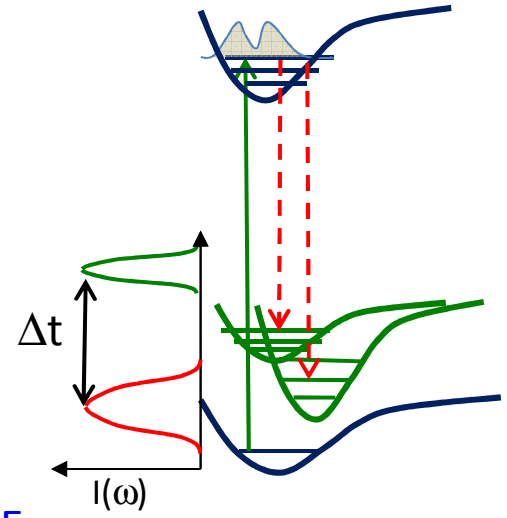


C. Weninger & N.R., Phys. Rev. A **88**, 053421 (2013)

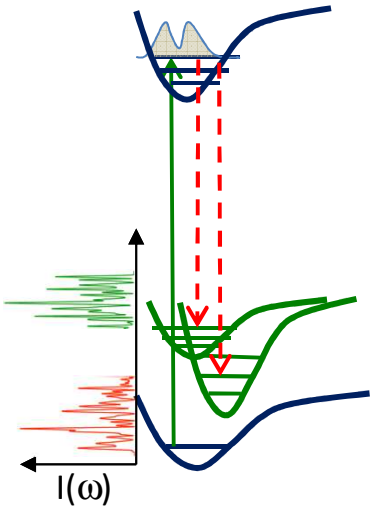
Different ways to for stimulated X-Ray Raman scattering



1. Two-color Raman scattering with seeded source
   Frequency-domain spectroscopy
   Pick 2 distinct frequencies within 10 eV SASE bandwidth allows selection of intermediate and final state



2. Impulsive Raman scattering with seeded source
   Pump-probe in spectroscopy
   short, transform limited pulses, of variable defined delay
   (2 eV bandwidth, pulse separation of 1-20 fs)



3. Impulsive Raman with broadband SASE
   Mixed time/frequency domain spectroscopy

in a statistical sense:
energy resolution – spectral coherence of SASE
temporal resolution - coherence time of source / pulse duration

Summary and Outlook

Stimulated x-ray emission processes are accessible at XFELS

1st demonstration of stimulated electronic x-ray Raman scattering in neon

Transfer of stimulated emission processes to molecules (gas phase):
2 upcoming experiments in 2014/2015 in collaboration with

- J.E. Rubensson, J. Nordgren, R. Faifel (Uppsala University)
- J. Küpper, T. Mullins, O. Mücke, F. Kārtner (CFEL)
- R. Coffee, J. Bozek (SLAC)
- A. Föhlisch, M. Beye (Helmholtz Zentrum Berlin)

Challenges

Spectral stability of x-ray pulses (SASE, self-seeded, seeded FELs) will determine the necessary gain regime (strong Raman gain for SASE, low-gain for seeded FELs)

Spectrally instable pulses require optically dense sample - difficult to interpret spectra!

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

Parameters Wish List

	Day 0	Nice to have
Experimental techniques	sRIXS, x-ray pump-probe techniques	
Source properties		
Energy range	280 eV (C Kedge) – 2.5 keV (K edges of P,S)	
Pulse duration	2 fs – 100 fs	0.1fs -100 fs
bandwidth	variable: 50 meV (seeded)	transform limited
Device properties		
Maximum Temporal delay	100 fs	1-2 ps (liquids)
Pulse intensity ratio	1:1 to 1:100 – variable!	
2 Colors	yes	
Symmetric delay around t=0	yes, for coherent control (STIRAP)	
Spatial separation behind sample	yes, different angle of incidence, allows to measure incoming/transmitted spectrum	different angles of incidence for two-pulse options
Add your suggestions	pump probe delay step size 0.1 fs	two-pulse mode, with one short, broadband pulse (impulsive Raman, for wavepacket creation), and second pulse should be narrow-bandwidth, long pulse (full intensity not necessary)