The Need for an Open Quantum System Approach to Describe High-Intensity X-ray Interactions with Matter

August 3rd, 2010
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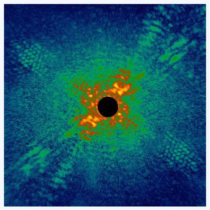
This work performed under the auspices of the U.S. Department of Energy
by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

LLNL-PRES-448504

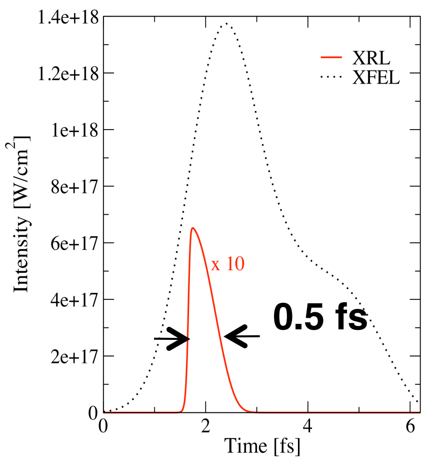
Non-linear optical physics in the X-ray regime



Linear effects (one-photon one-electron interactions):
- Coherent diffractive imaging
- Time-resolved photo-absorption spectroscopy (RIXS, XANES)
- Time-resolved photoelectron spectroscopy
- Spectroscopy of highly-charged ions (EBIT)

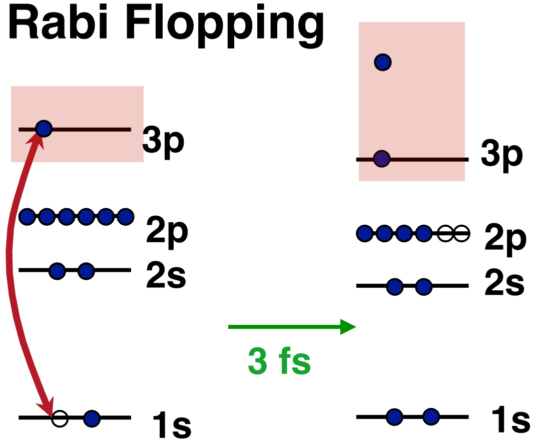
Quantum optics transferred to the X-ray regime

Atomic x-ray lasing with inner-shell transitions



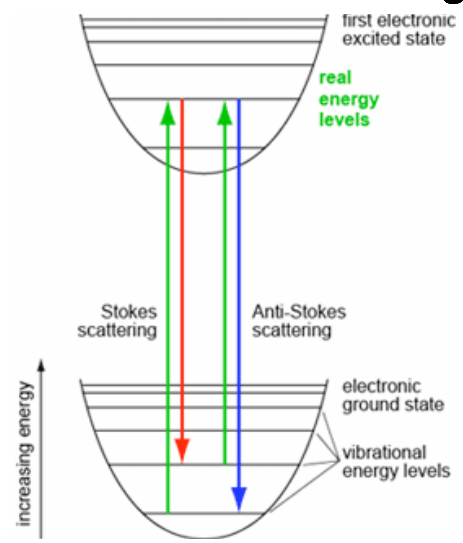
Rohringer and London, Phys. Rev. A (2010)

Rabi Flopping

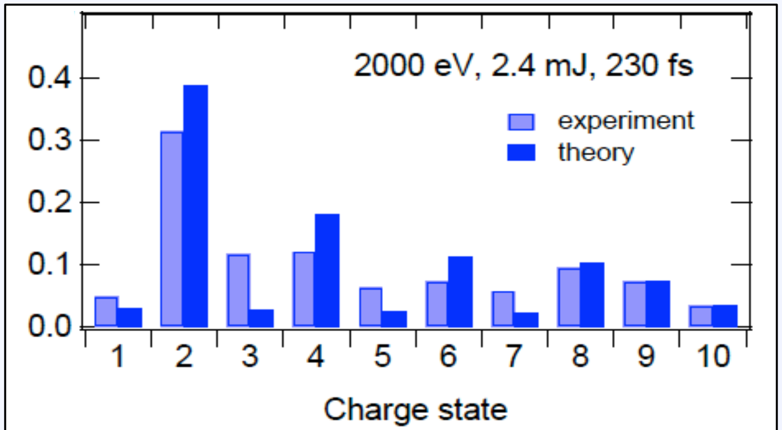


Rohringer and Santra, Phys. Rev. A (2008).

Resonant Raman Scattering

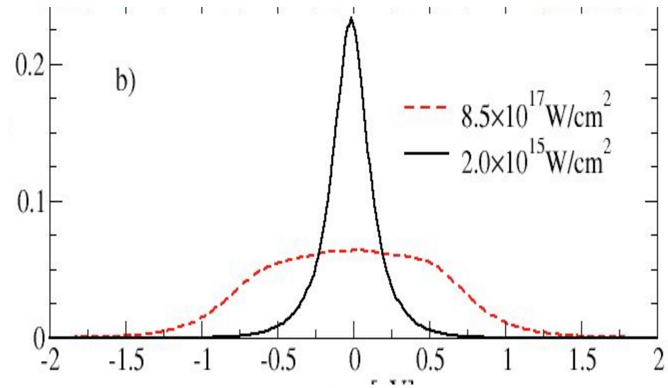


Non-resonant versus resonant high-intensity x-ray matter interaction



L. Young et al., Nature 466, 56 (2010)

Resonant Auger-electron spectrum



Rohringer and Santra,
Phys. Rev. A (2008)

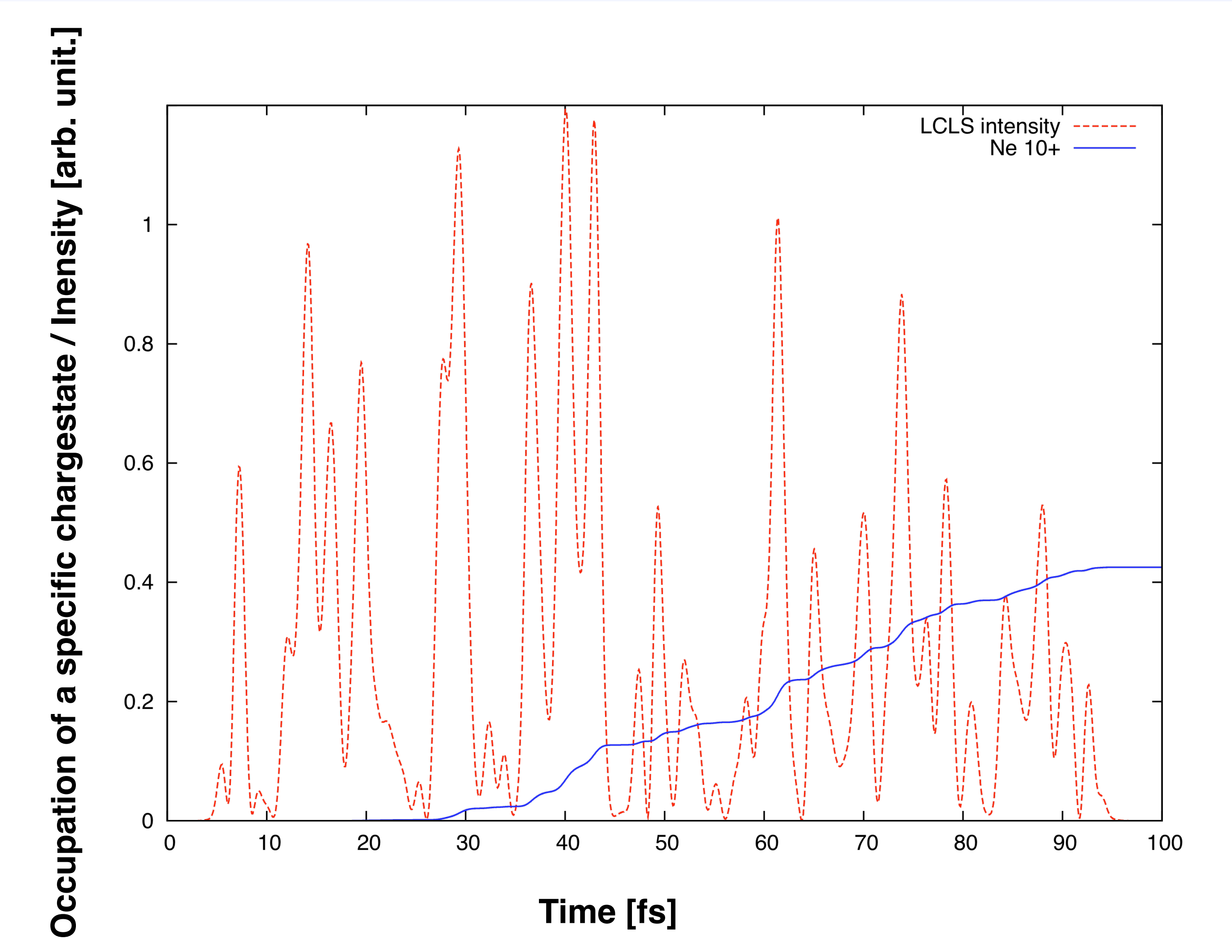
Non-resonant interaction

- **1st user experiment at LCLS on Neon**
- Sequence of one-photon absorption events followed by Auger or radiative decay
- Model of **kinetic rate equations valid**

Resonant interaction

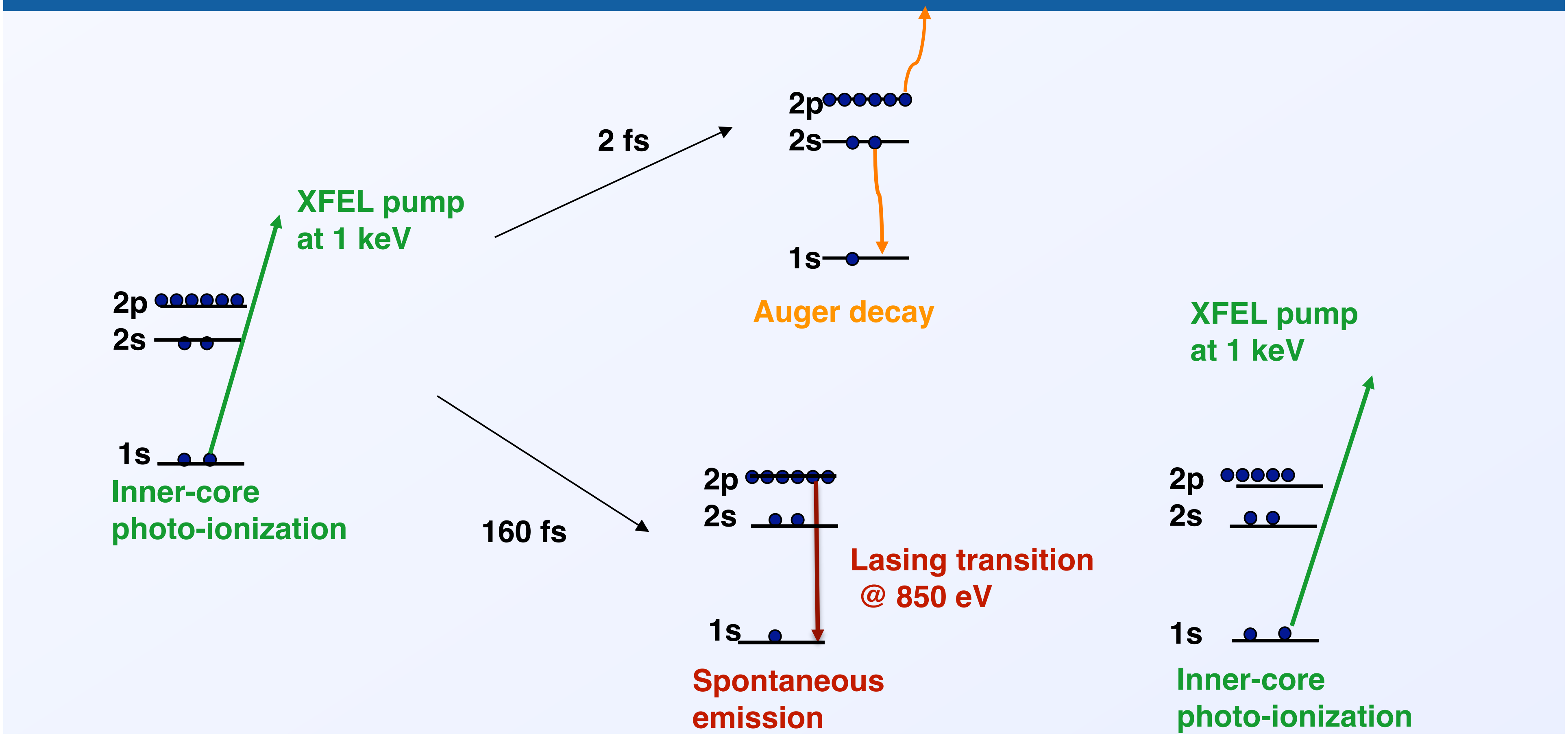
- **Increased coupling strength on resonance**
- Perturbation theory breaks down
- **Non-linear** description necessary
- **Optical Bloch equations** for a **dissipative system**

Focusing LCLS into a gas sample of Neon
Parameters: pulse of 100 fs, 10^{12} photons, ω=1.4 keV, focused to 2 μm

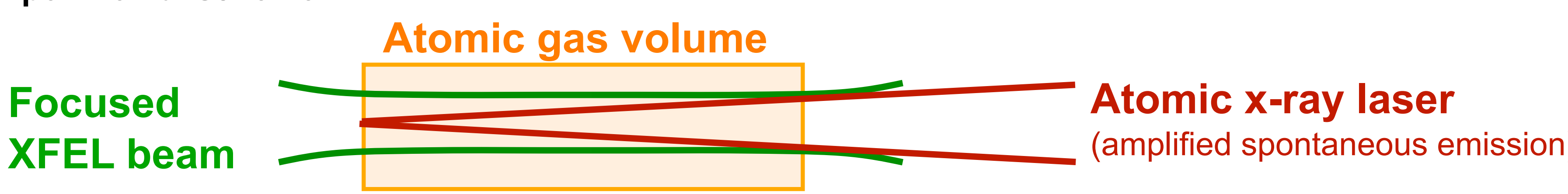


Rohringer and Santra, Phys. Rev. A, (2008).

An atomic inner-shell X-ray laser pumped with XFEL radiation based on Neon

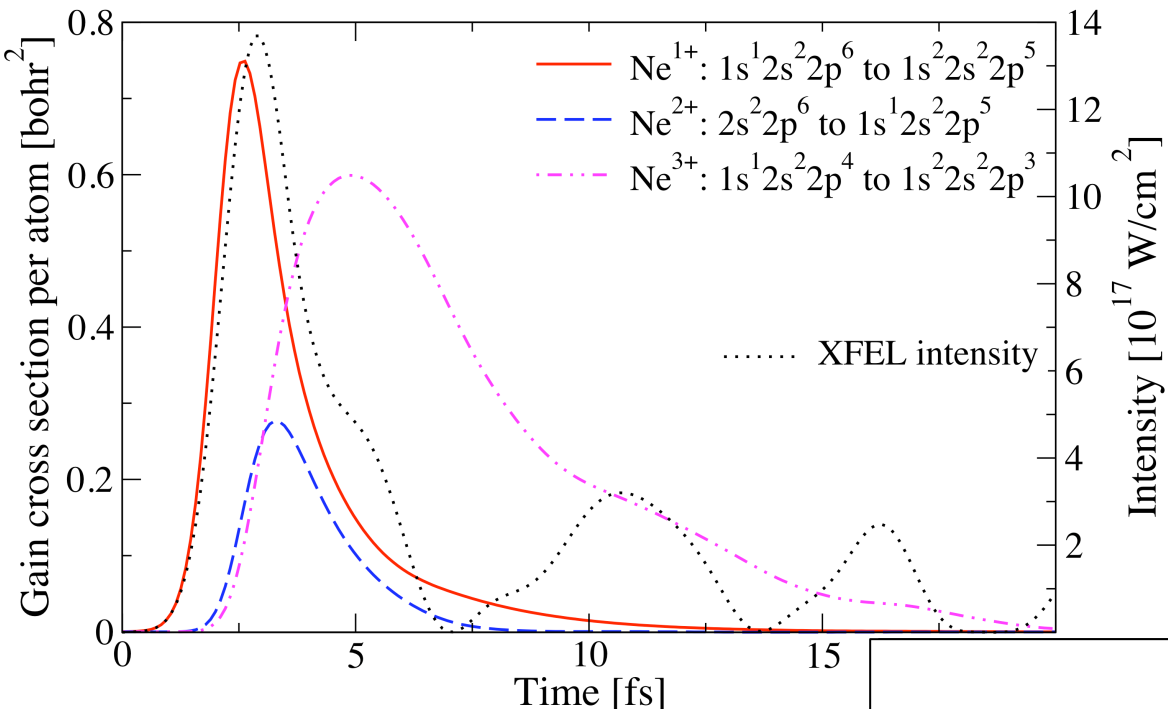


Experimental scheme:



Small-gain parameters at 1 keV pumping energy (single atom calculcation)

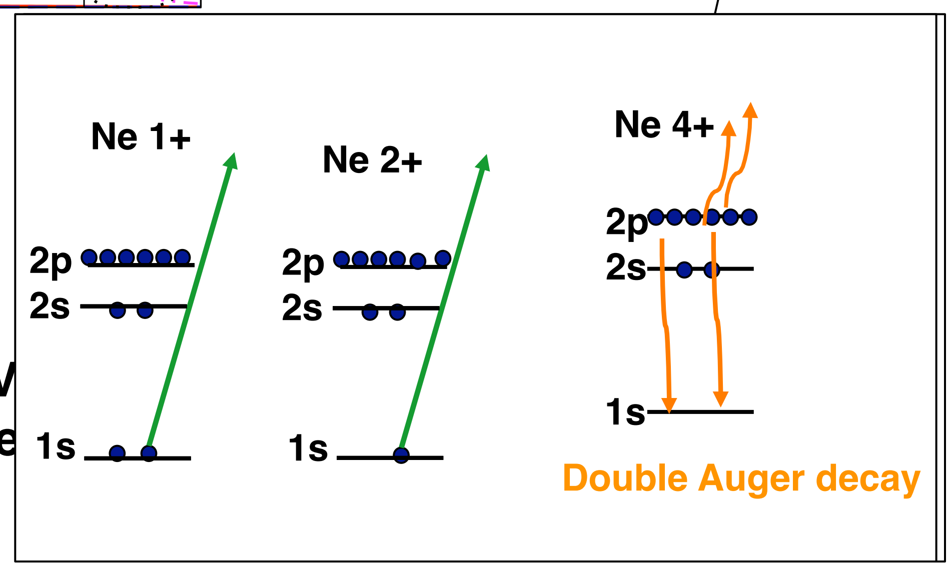
$$I(z,t) = I(0,t) \cdot e^{g \cdot n \cdot z}$$



$$\Delta\omega/\omega = 3 \cdot 10^{-4}$$

$$g(t) = n_U(t)\sigma_{stim} - n_L(t)\sigma_{abs}$$

Electron–ion collision times: at 100 eV
(for density of 10^{19} cm^{-3}) at 800 e
ions are at room temperature !



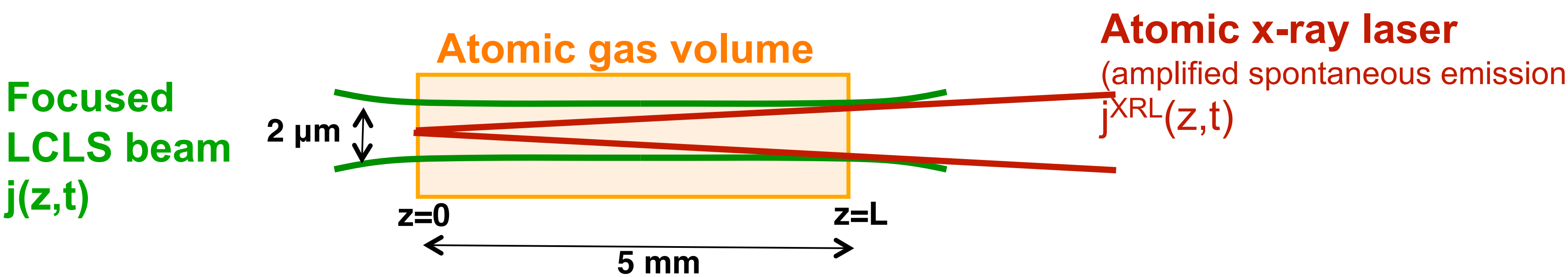
One-dimensional self-consistent gain calculation

Upper lasing level:

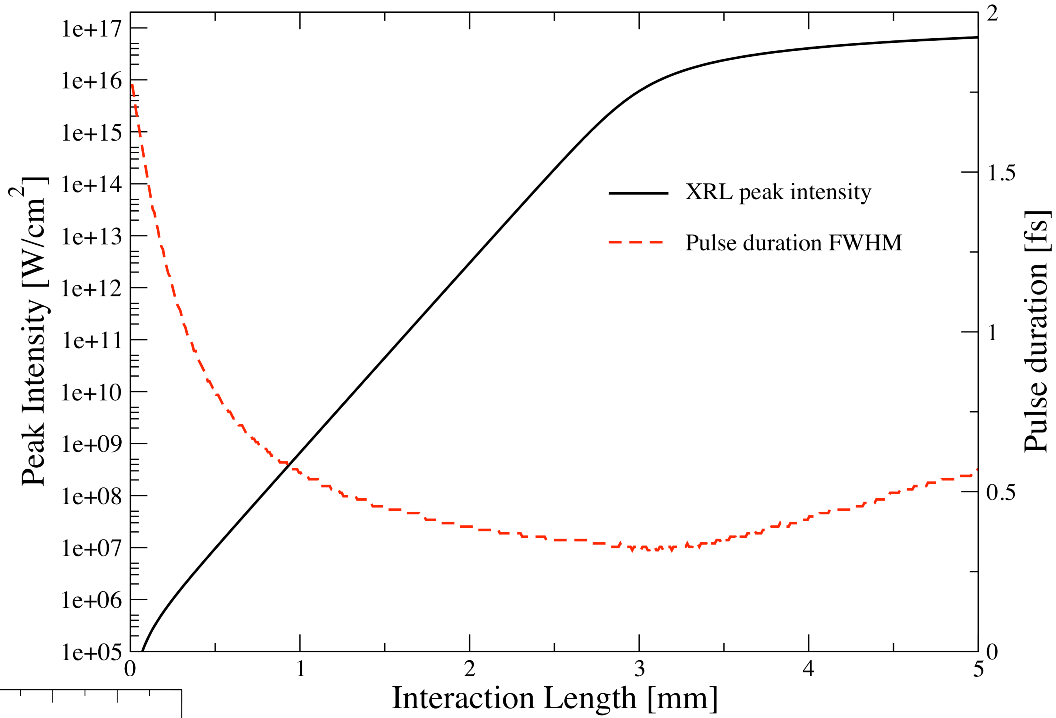
$$\frac{dN_U(z,t)}{dt} = \sum \sigma_i^v j(\tilde{t}_z) N_i^S(\tilde{t}_z) + \sum \sigma_i^c j(\tilde{t}_z) N_i^S(\tilde{t}_z)$$
$$-\sigma^{se}\left[j_+^{XRL}(z,t) + j_-^{XRL}(z,t)\right] N_U(z,t)$$
$$+\sigma^{abs}\left[j_+^{XRL}(z,t) + j_-^{XRL}(z,t)\right] N_L(z,t)$$
$$-\left[A_{U\to L} + p_U^A + (\sigma_U^v + \sigma_U^c) j(\tilde{t}_z)\right] N_U(z,t)$$

X-ray laser flux in forward and backward direction:

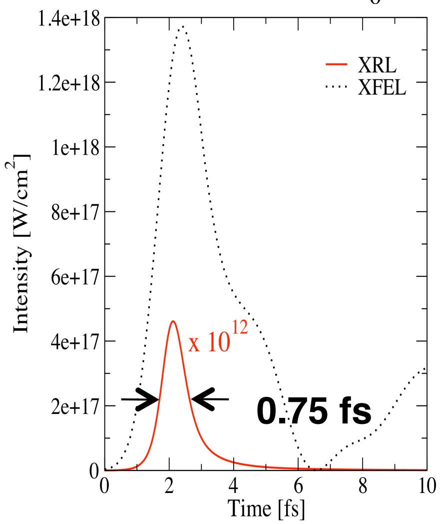
$$\frac{dj_\pm^{XRL}}{dt} = j_+^{XRL}(z,t) c n_A \left[\sigma^{se} N_U(z,t) - \sigma^{abs} N_L(z,t)\right]$$
$$+\frac{\theta_\pm(z)}{4\pi} A_{U\to L} N_U(z,t) n_A c \mp c \frac{dj_\pm^{XRL}}{dz}$$

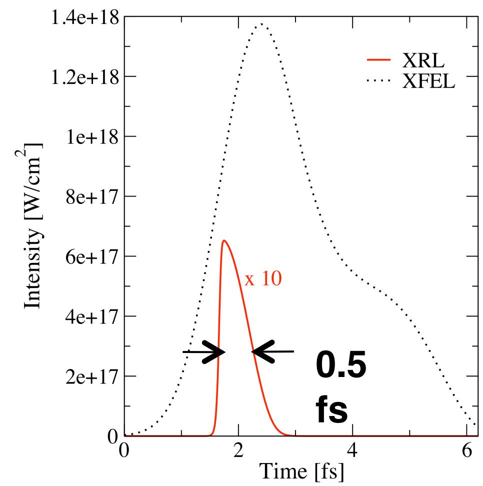


Expected output of atomic inner-shell x-ray laser



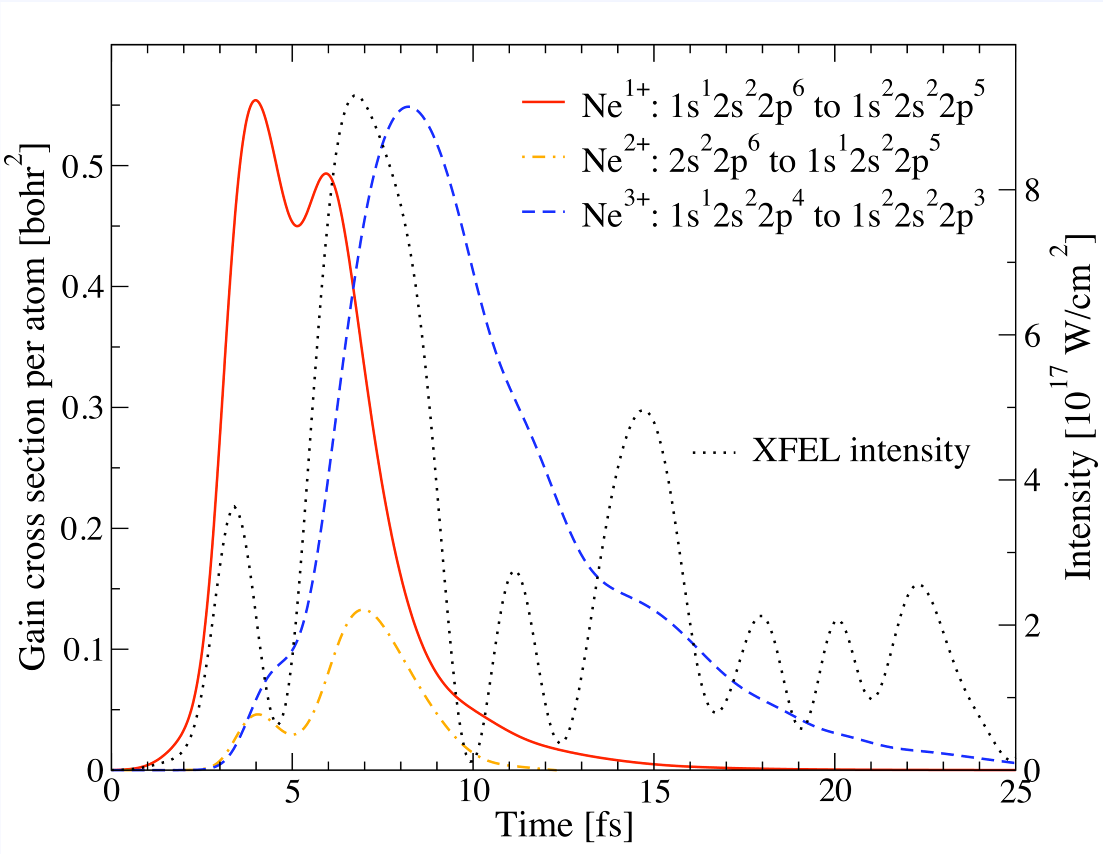
9×10^9 photons in XRL line
Intensity: 7×10^{16} W/cm^2
Duration: 0.5 fs





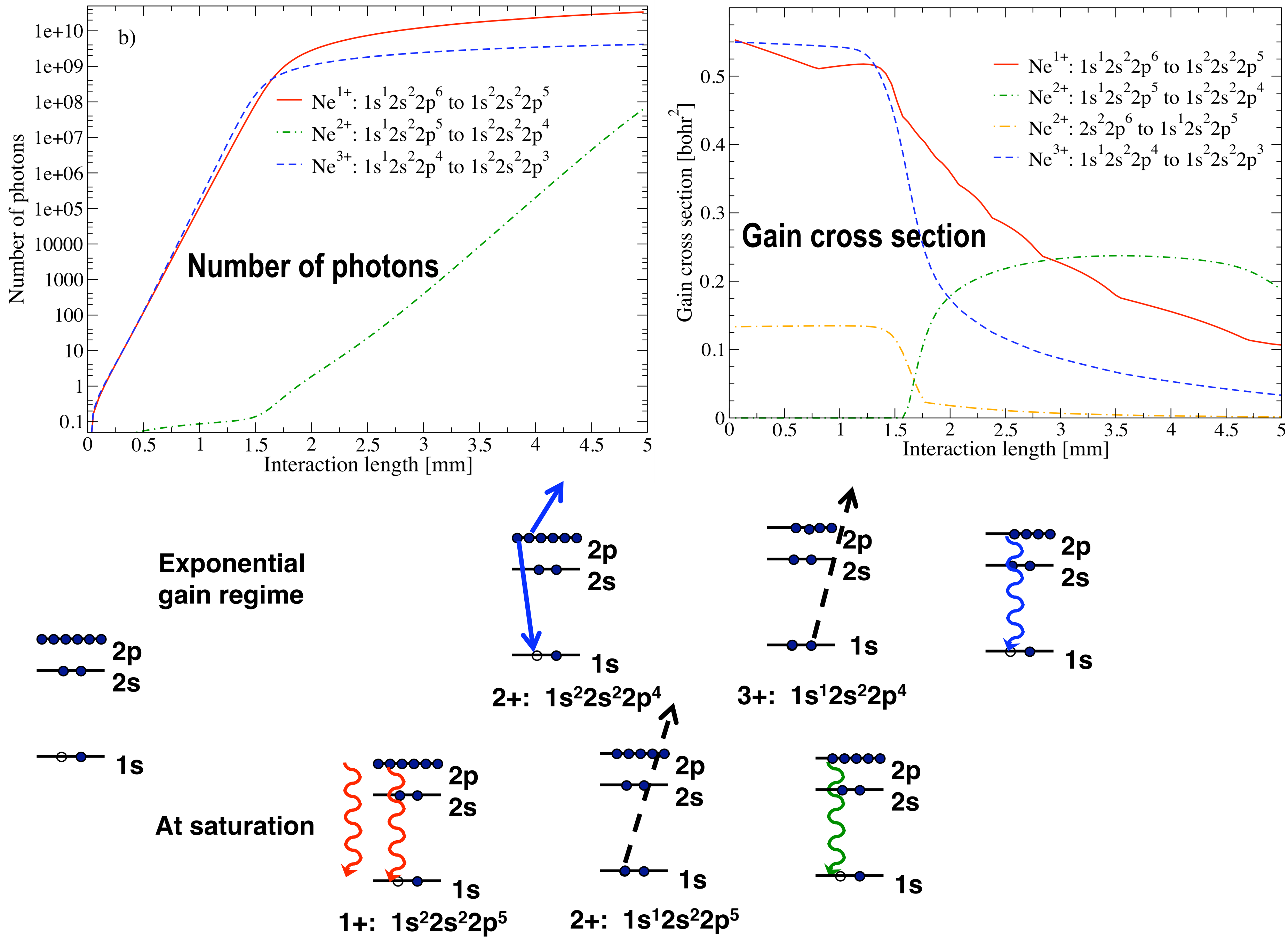
$n = 4 \times 10^{18}$ cm^{-3}, LCLS: 100 fs, 10^{12} photons per pulse, focal diameter 2 μm

Saturation of more than one lasing line seems possible



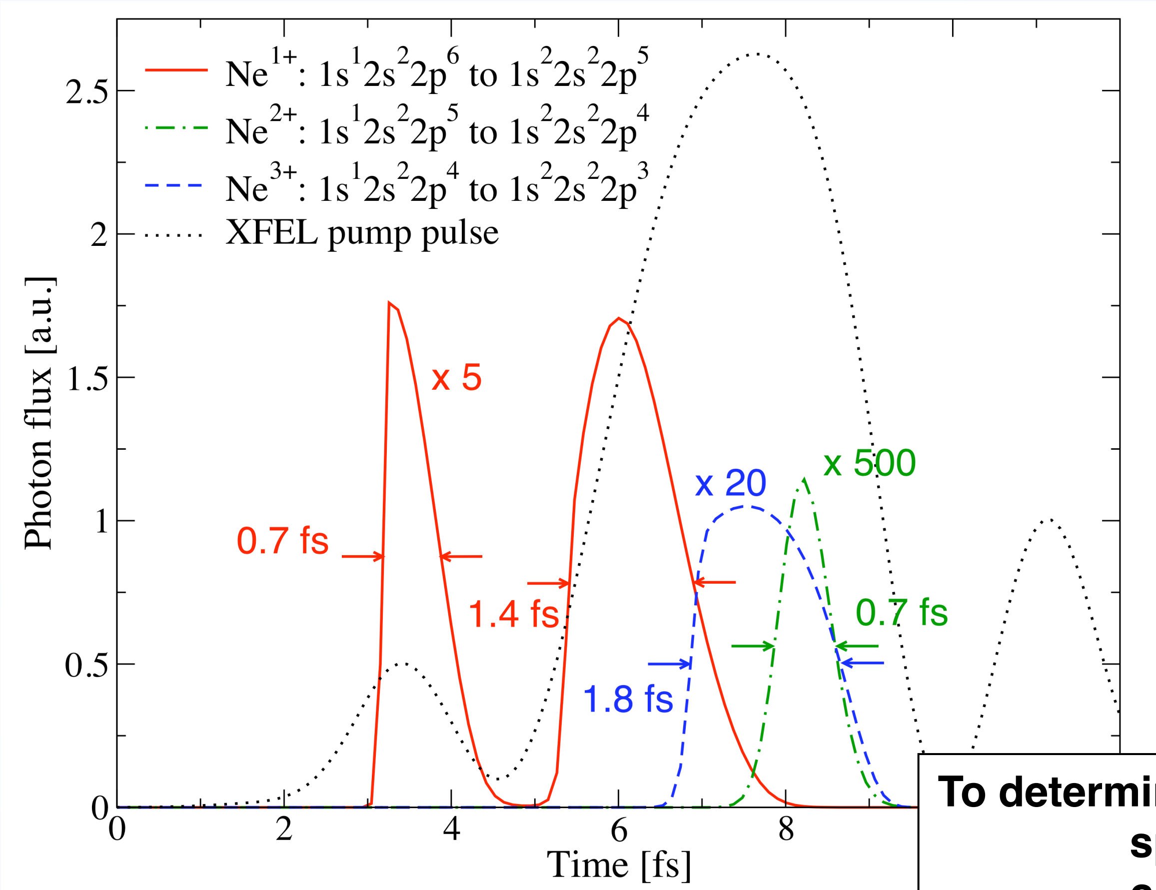
Rohringer and London, Phys. Rev. A (2009).

XFEL: 2 µm focus, 5×10^{12} photons per pulse, 100 fs pulse, 1keV energy
gas density: 1×10^{19} atoms/cm^3



b)
Number of photons
Interaction length [mm]
Ne1+: 1s1 2s2 2p6 to 1s2 2s2 2p5
Ne2+: 1s1 2s2 2p5 to 1s2 2s2 2p4
Ne3+: 1s1 2s2 2p4 to 1s2 2s2 2p3
Gain cross section
Gain cross section [bohr2]
Ne2+: 2s2 2p6 to 1s1 2s2 2p5
Exponential gain regime
At saturation
2p
2s
1s
2+: 1s22s22p4
3+: 1s12s22p4
1+: 1s22s22p5
2+: 1s12s22p5

Output at end of amplifying plasma column for 1 keV pump
Pathway to multi-color x-ray fs pump-probe experiments

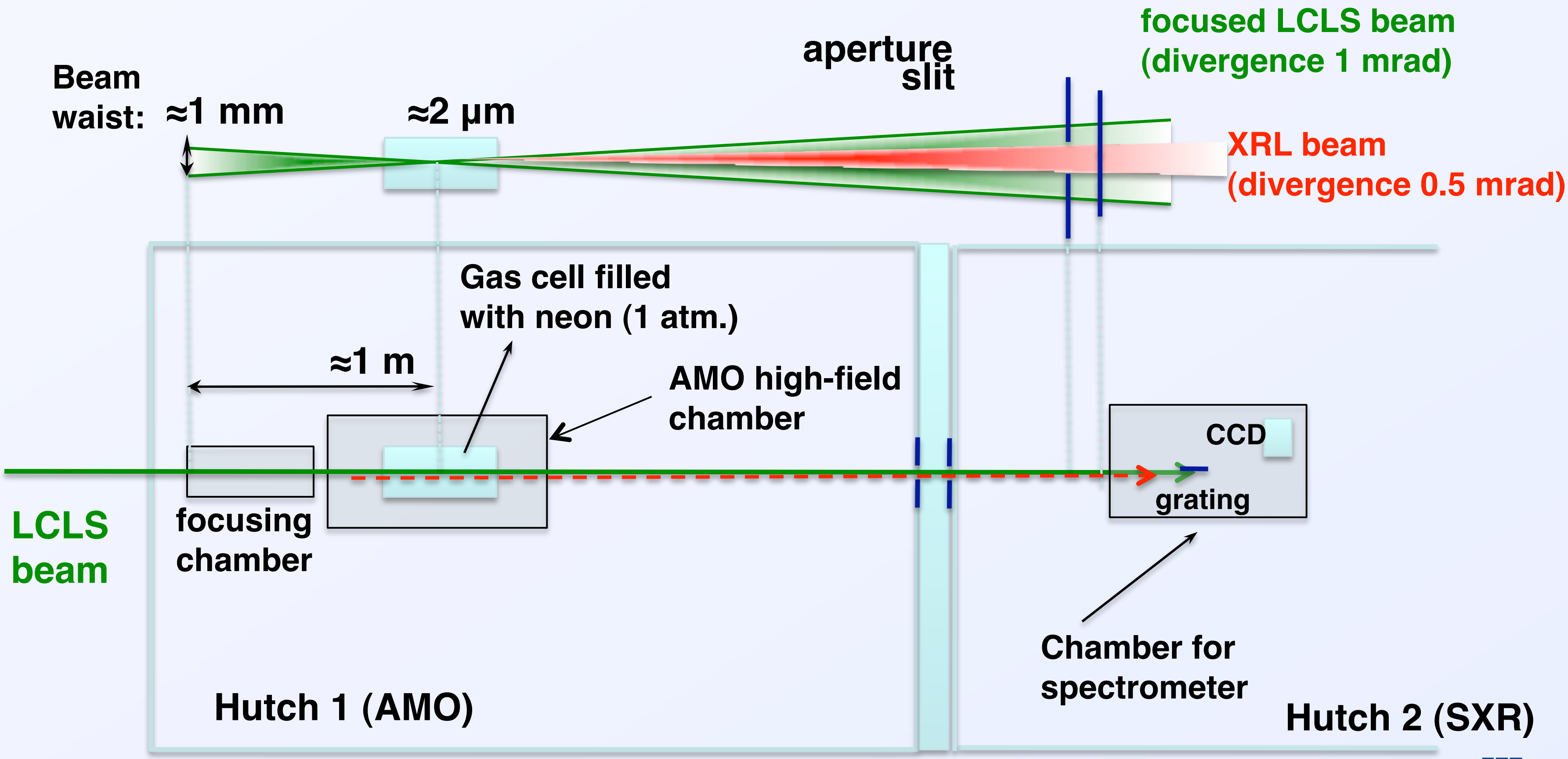


E	#(photons)	I_{peak}
850 eV	$3x10^{10}$	$7x10^{16}$ W/cm^2
862 eV	$4x10^9$	$1x10^{16}$W/cm^2
855 eV	$6x10^7$	$5x10^{14}$W/cm^2

To determine
spectrum,
coherence properties,
propagation effects,
refraction, etc.
-> Solve coupled Maxwell-Bloch equations

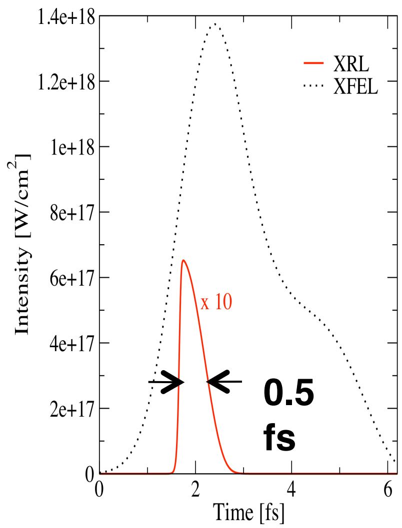
Scheduled experiment of XRL lasing scheme in Sept. 2010

Photo-ionization pumping scheme at different wavelengths

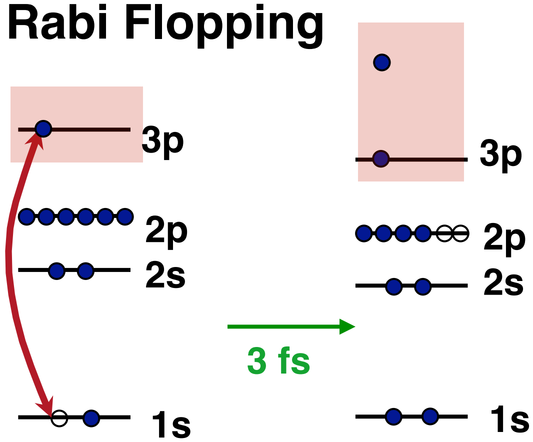


Non-linear optical physics in the X-ray regime

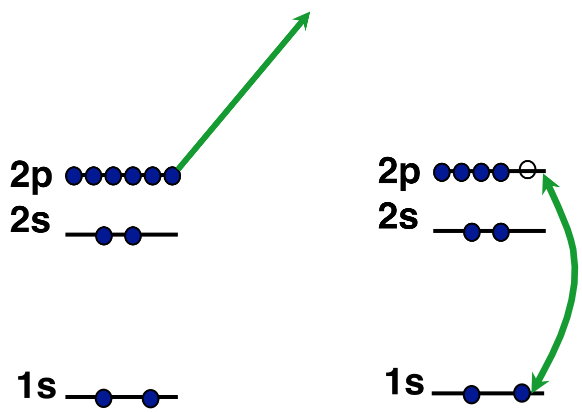
Atomic x-ray lasing with inner-shell transitions



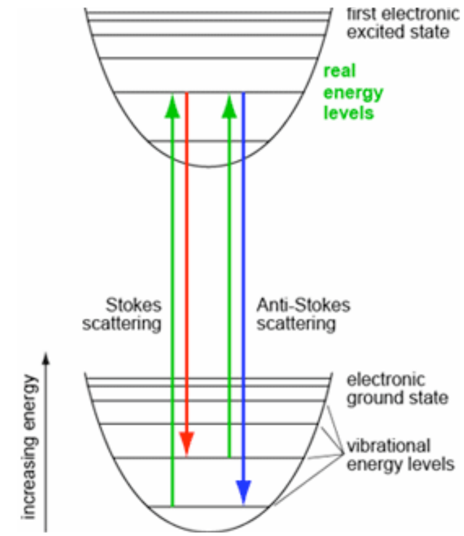
Rabi Flopping



Rohringer and Santra, Phys. Rev. A (2008).



Resonant Raman Scattering

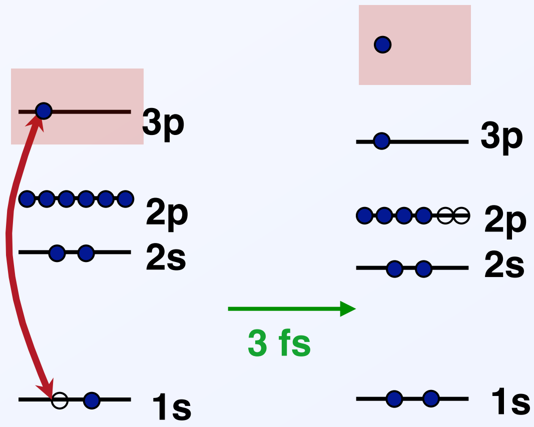


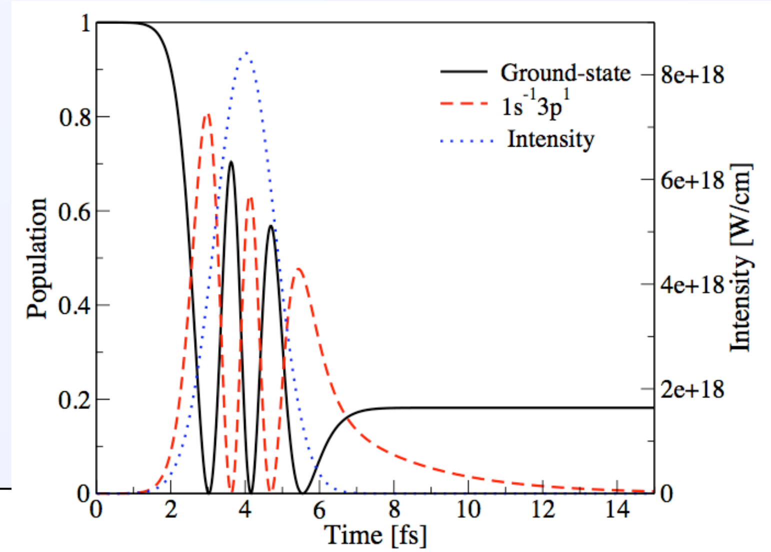
Resonant Auger effect at high X-ray intensities, Ne 1s-3p transition, Wave-function approach

Coupling strength (Rabi frequency)

$\Omega(t) = |E(t) \cdot \langle 3p|z|1s\rangle|$

$|\langle 3p|z|1s\rangle| \approx 0.01$ a.u.





$$|\Psi, t\rangle = \tilde{c}_1(t)|1\rangle + \tilde{c}_2(t)|2\rangle + \sum_i \int d\epsilon_i \tilde{g}_i(\epsilon_i, t)|i, \epsilon_i\rangle$$

$$i\dot{c}_1(t) = -\frac{\delta}{2}c_1(t) + \frac{\mathcal{R}^*(t)}{2}c_2(t)$$

$$i\dot{c}_2(t) = -i\frac{\Gamma_{1s^{-1}}}{2}c_2(t) + \frac{\delta}{2}c_2(t) + \frac{\mathcal{R}(t)}{2}c_1(t)$$

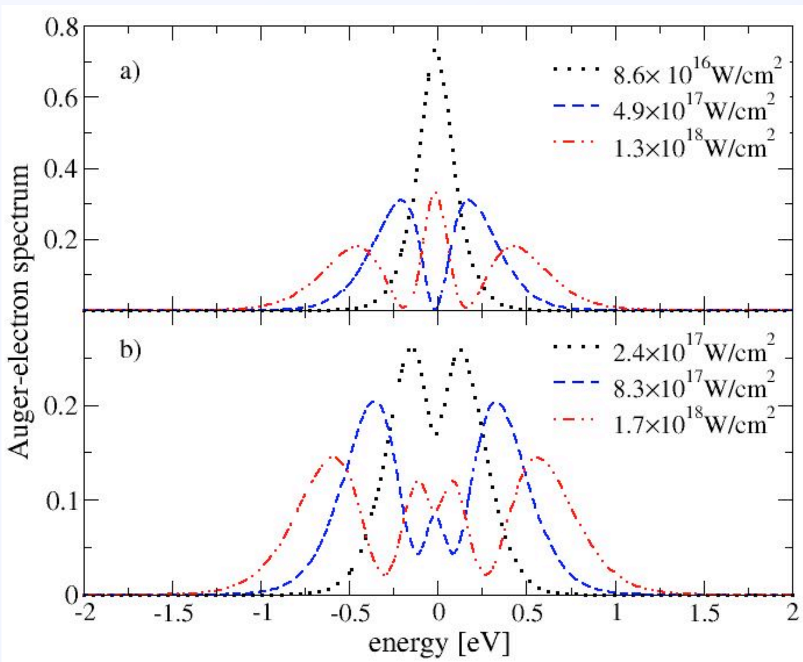
$$i\dot{g}_i(\epsilon_i, t) = \left[E_i^{(+)} + \epsilon_i - E_2 + \frac{\delta}{2}\right]g_i(\epsilon_i, t) + \sqrt{\frac{\Gamma_i}{2\pi}}c_2(t)$$

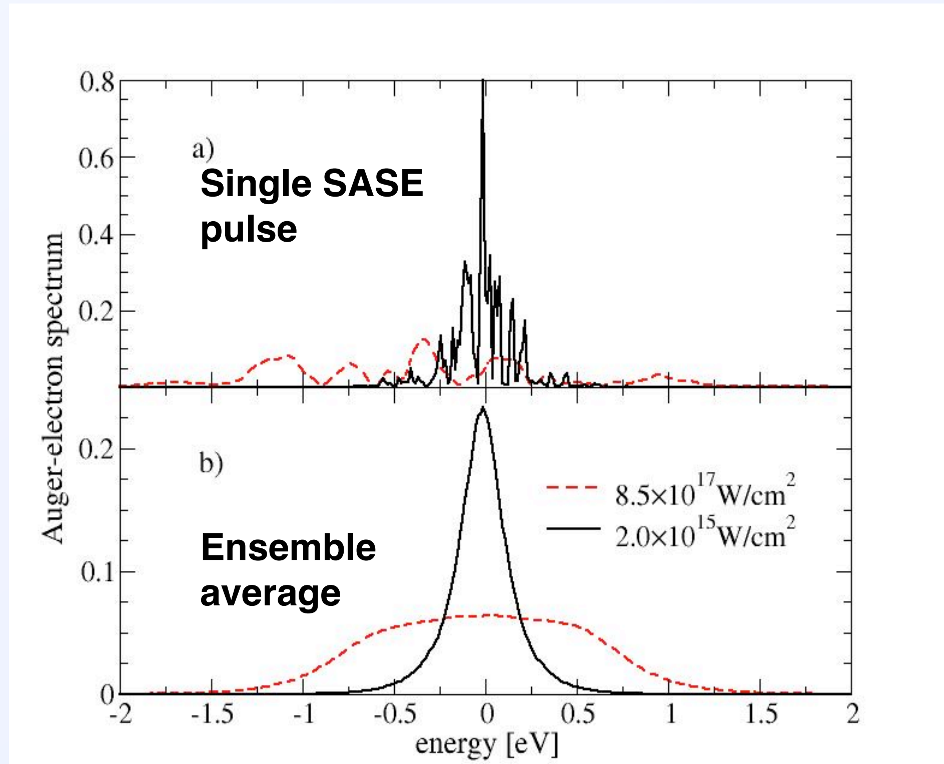
$$P_i(\epsilon_i) = \lim_{t\to\infty}|g_i(\epsilon_i, t)|^2$$
$$= \frac{\Gamma_i}{2\pi}\lim_{t\to\infty}\left|\int_{-\infty}^{t} dt' c_2(t') e^{i[E_i^{(+)}+\epsilon_i - E_2 + \delta/2]t'}\right|^2$$

Rohringer and Santra, Phys. Rev. A (2008)

Resonant Auger-electron spectrum gets broadened

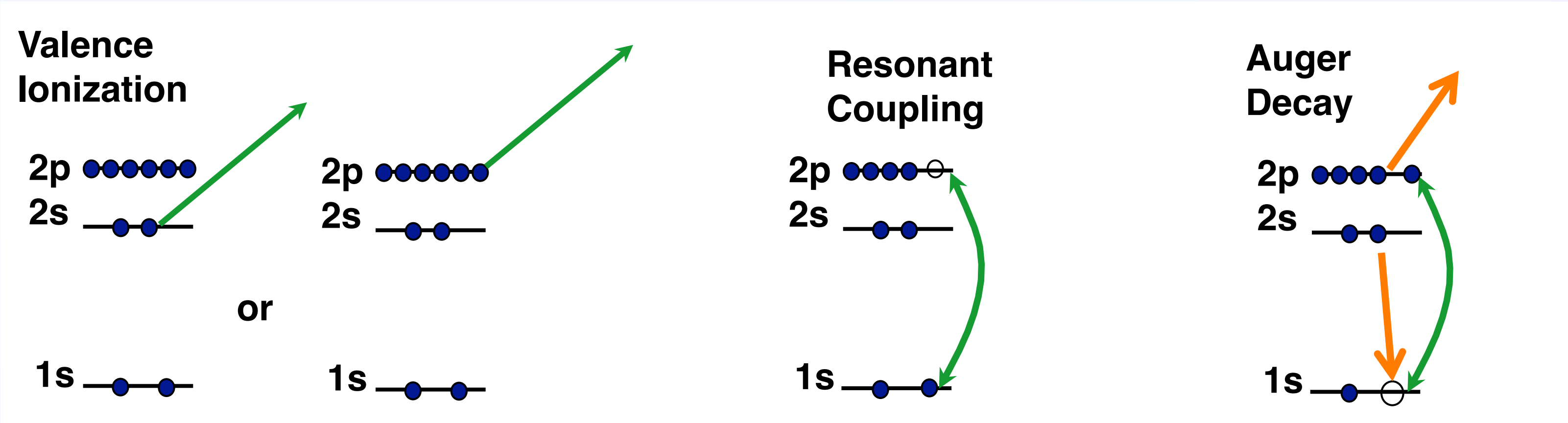
Coherent Gaussian Pulse





Rohringer and Santra, Phys. Rev. A (2008)

Generalized Bloch equations for open quantum system
Calculate time-dependent ionic density matrix



Loss of coherence, Open Quantum System, Unobserved photo electrons

$$\dot{\rho}_{11} = \left[ie^{-i\delta t} \rho_{21} \frac{R^*(t)}{2} + cc \right] + \sigma_1 j(t) p_0(t)$$

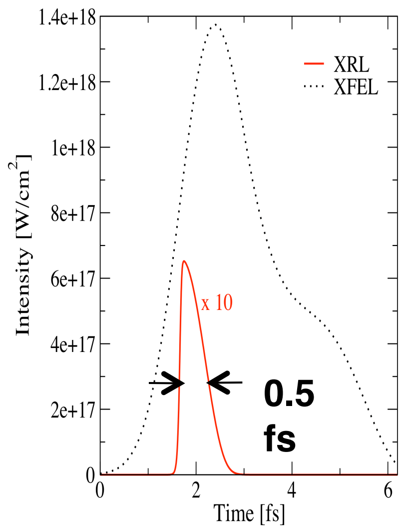
$$\dot{\rho}_{22} = -\Gamma_2 \rho_{22} + \left[ie^{i\delta t} \rho_{12} \frac{R(t)}{2} + cc \right]$$

$$\dot{\rho}_{12} = -\frac{\Gamma_2}{2} \rho_{12} + ie^{-i\delta t} (\rho_{22} - \rho_{11}) \frac{R^*(t)}{2}$$

Rohringer and Santra, work in progress

Self-Stimulated Resonant Raman Scattering

Atomic x-ray lasing with inner-shell transitions



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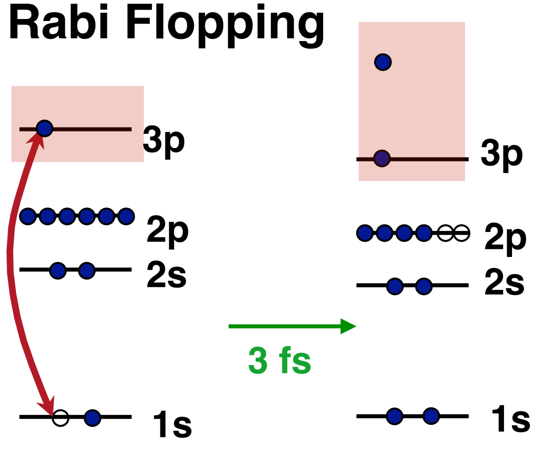

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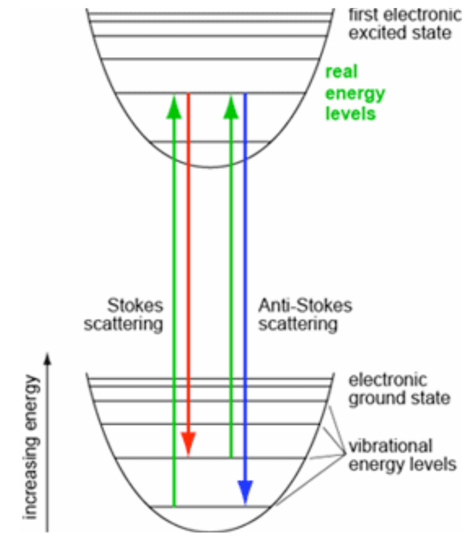
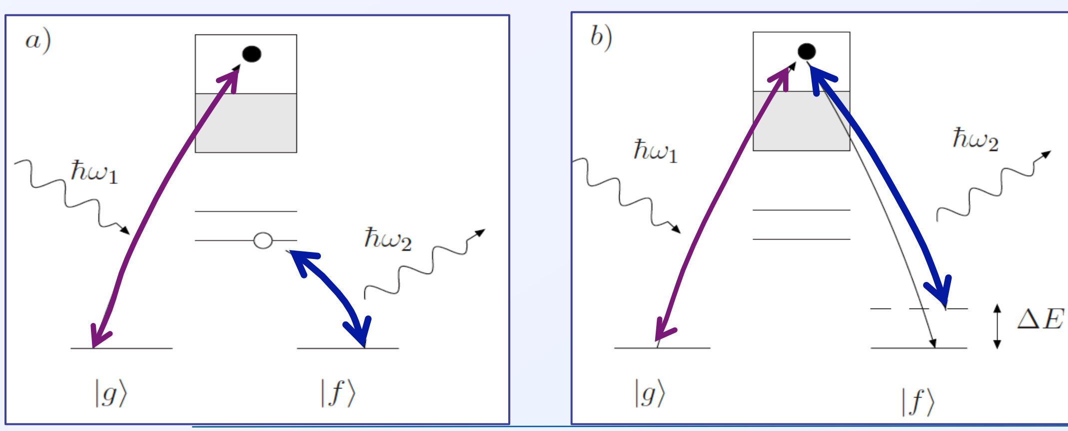
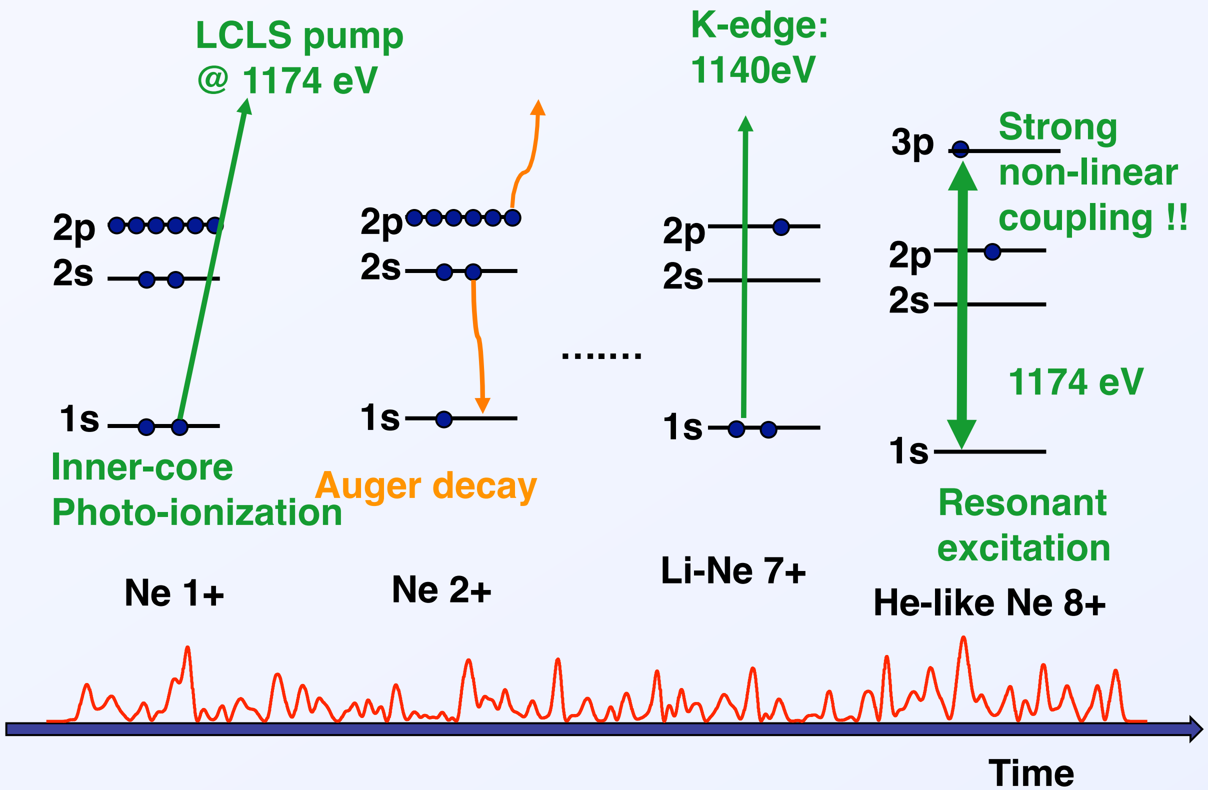
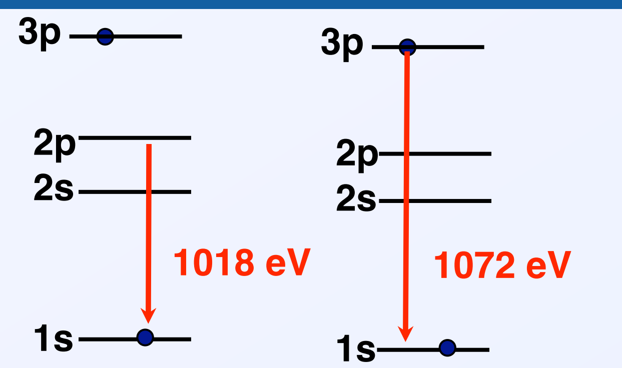


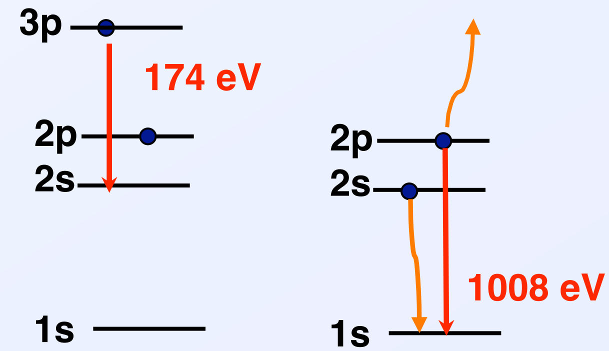


Proposed experiment: Resonant pumping scheme





Pathway will depend
On x-ray intensity !
Comparable oscillator strengths!



Self-stimulated resonant Raman scattering

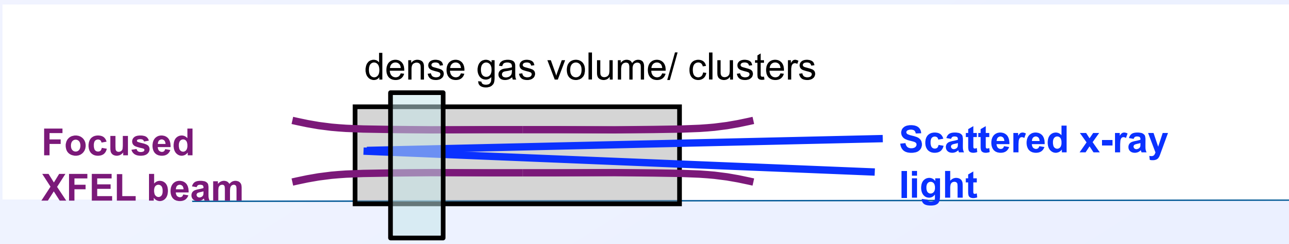
Proposal: Stochastic wave function approach

Adopt stochastic wave-function approach from quantum optics

Ionic reduced density matrix sampled by ensemble of **pure states**
Ionic wave function evolves **deterministic** (resonant interaction with XFEL)
+ random (stochastic) **quantum jumps** to simulate
-photo ionization processes
-Auger decay
-eventually treat electron impact ionization, excitation, de-phasing

Quantum jump approach:
Jump operators: derived by semiclassical approximation ?

Test on single-atom case
by comparing to generalized optical Bloch equations
(equivalent descriptions of determining ionic reduced density matrix)



Acknowledgements

LLNL: Richard London
James Dunn
Felicie Albert
Sebastien Le Pape
Alexander Graf

CSU: Jorge Rocca
Duncan Ryan
Mike Purvis

SLAC: John Bozek
Christoph Bostedt

CFEL: Robin Santra

We acknowledge support of this project by
LLNL's LDRD (Laboratory Directed Research and Development) program.