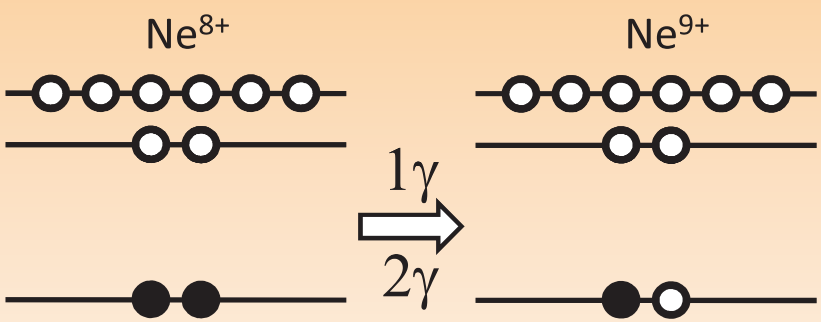
Nonlinear atomic response to ultraintense and ultrashort x-ray pulses



Sang-Kil Son

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CFEL SCIENCE
Center for Free-Electron Laser Science
CFEL is a scientific cooperation of the three organizations:
DESY – Max Planck Society – University of Hamburg

DESY

Author list

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Nonlinear Atomic Response to Intense Ultrashort X Rays

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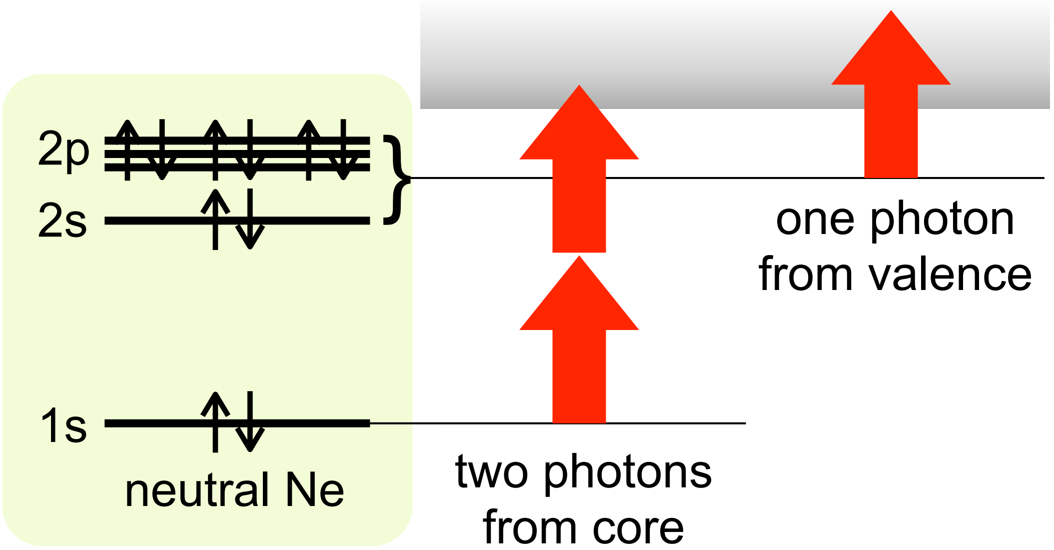
Nonlinear response in the x-ray regime

- Rapid decrease of two-photon absorption cross section with increasing frequency ν

$$\sigma^{(2)} \sim \sigma^{(1)}\, \nu^{-1}\, \sigma^{(1')}$$

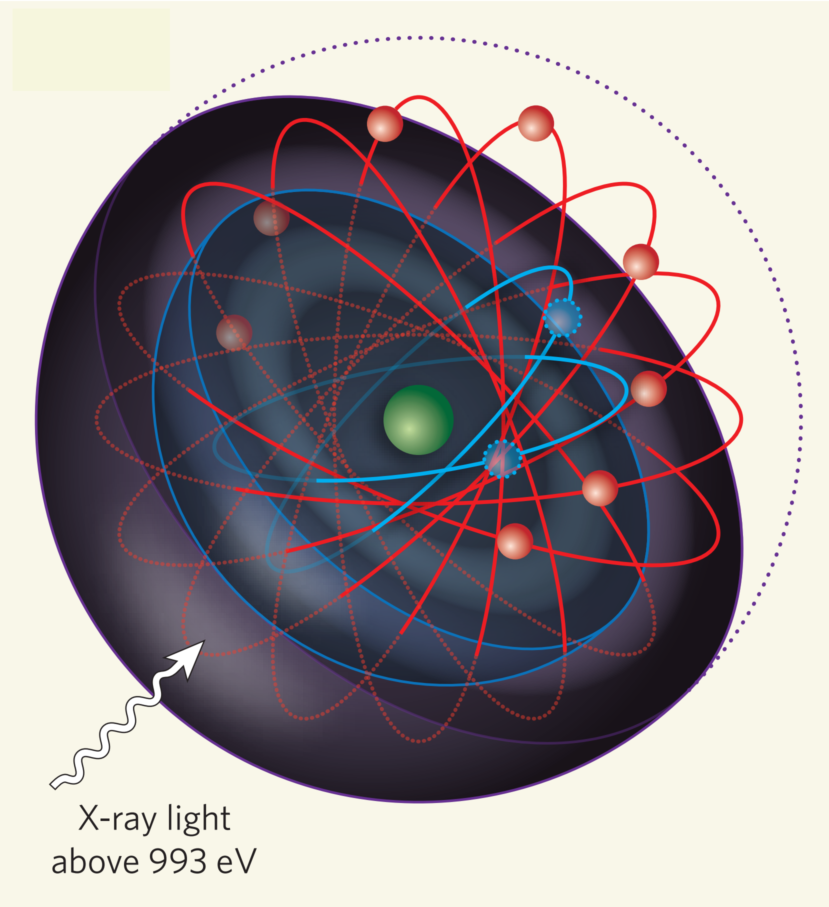
→ High intensity needed: X-ray FEL (free-electron laser)

- Linear response from valence-shell electrons, rather than nonlinear response from core-shell electrons



→ Use ions (Ne^{8+}) without any valence electrons

Ne^{8+} production at ultraintense x-rays



Picture from Nature **466**, 35 (2010)

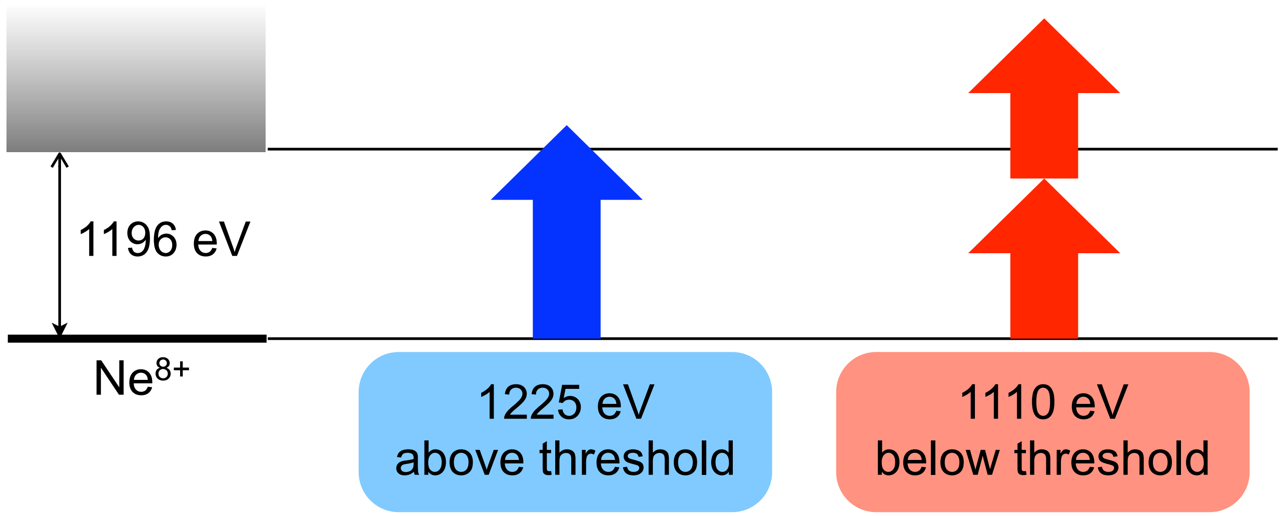
- The first LCLS experiment [Young *et al.*, Nature **466**, 56 (2010)] revealed multiphoton ionization mechanisms in ultraintense x-ray pulses.
- Many electrons in Ne can be stripped off through a series of one-photon absorption and relaxation (Auger decay).

↓

Production of Ne^{8+}

Experimental setup

- Conducted at LCLS (Linac Coherent Light Source) X-ray FEL: 0.8–2 keV with a peak intensity ~10^{17} W/cm^2 and 100 fs pulse duration
- Choice of a photon energy below / above K-shell threshold of Ne^{8+}



Theoretical model

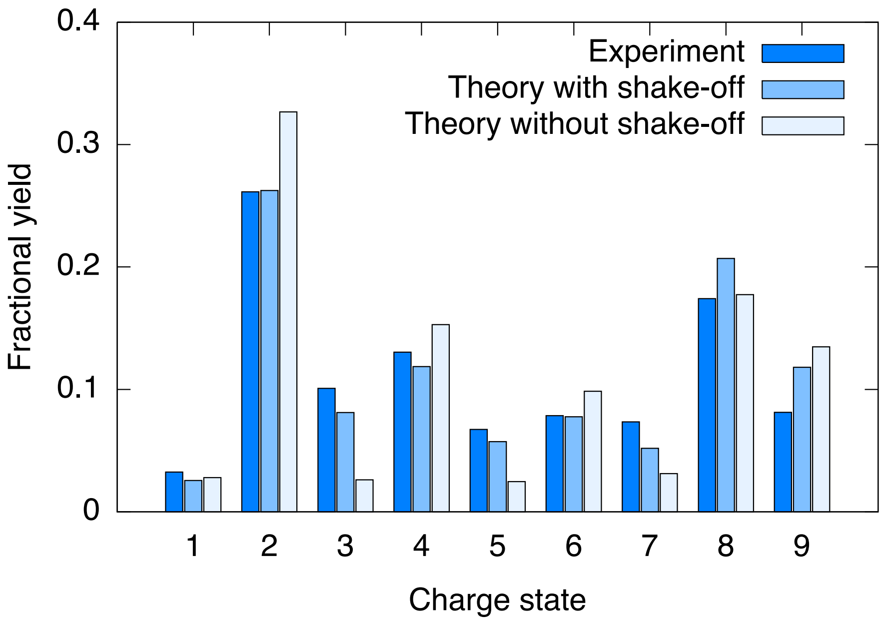
- XATOM: integrated toolkit for X-ray atomic physics [PRA **83**, 033402 (2011)]
- Photoionization $$\sigma_{\mathrm{P}}(i,\omega) = \frac{4}{3}\alpha\pi^2\omega N_i \sum_{l_j=|l_i-1|}^{l_i+1} \frac{l_>}{2l_i+1} \left| \int_0^\infty P_{n_i l_i}(r) P_{\varepsilon l_j}(r)\, r\, dr \right|^2$$
- Auger decay $$\Gamma_{\mathrm{A}}(i,jj') = \pi \frac{N_i^{\mathrm{H}} N_{jj'}}{2l_i+1} \sum_{L=|l_j-l_{j'}|}^{l_j+l_{j'}} \sum_{S=0}^{1} \sum_{l_{i'}} (2L+1)(2S+1)|M_{LS}(j,j',i,i')|^2$$
- Fluorescence $$\Gamma_{\mathrm{F}}(i,j) = \frac{4}{3}\alpha^3 (I_i - I_j)^3 \frac{N_i^{\mathrm{H}} N_j}{4l_j+2} \cdot \frac{l_>}{2l_i+1} \left| \int_0^\infty P_{n_i l_i}(r) P_{n_j l_j}(r)\, r\, dr \right|^2$$
- Shake-off process $$p_{\mathrm{S}}(i;I,I') = 1 - \left| \int_0^\infty P_{n_i l_i}(r;I) P_{n_i l_i}(r;I')\, dr \right|^2$$
- Calculate all above for all configurations (63 config. for Ne)
- Rate equation to simulate electronic damage dynamics

$$\frac{d}{dt} P_I(t) = \sum_{I' \neq I}^{\text{all config.}} \left[\Gamma_{I' \to I} P_{I'}(t) - \Gamma_{I \to I'} P_I(t) \right]$$

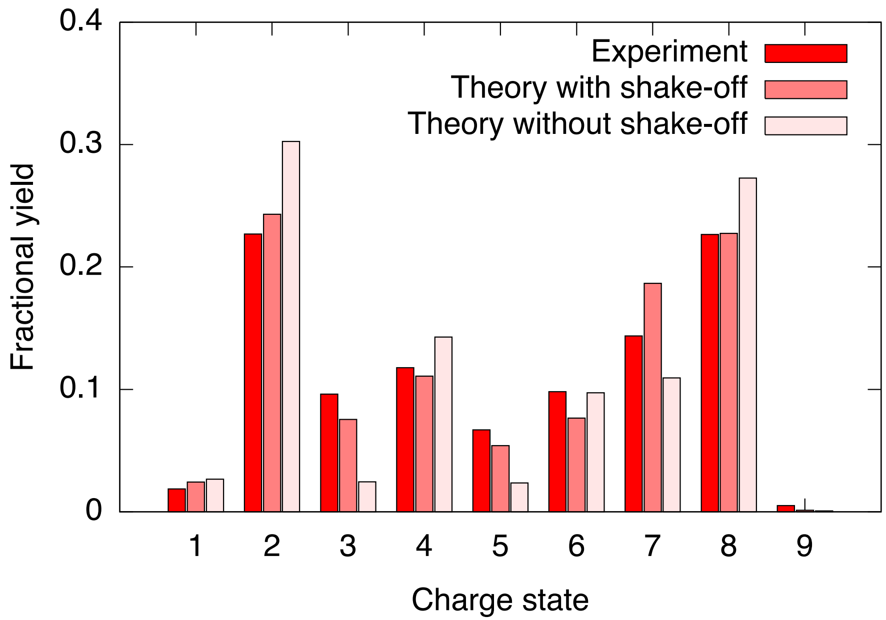
- Combined with coherent scattering ➔ **Poster A26.15 Thu 16:00 P2**

Charge state distributions of Ne

1225 eV: above threshold



1110 eV: below threshold



- Even / odd alternation and effect of shake-off processes
- Less abundance of Ne^{9+} at 1110 eV

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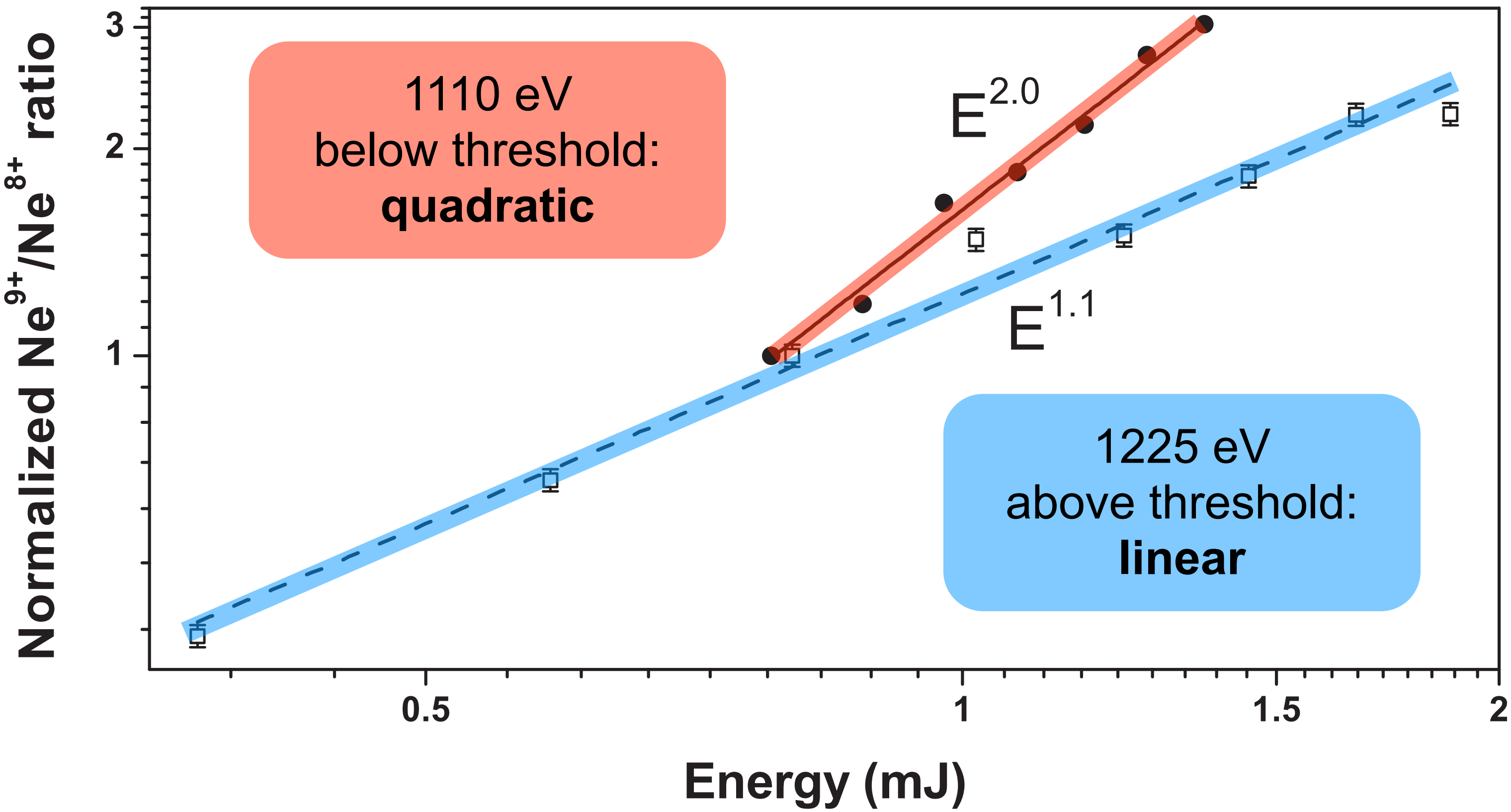
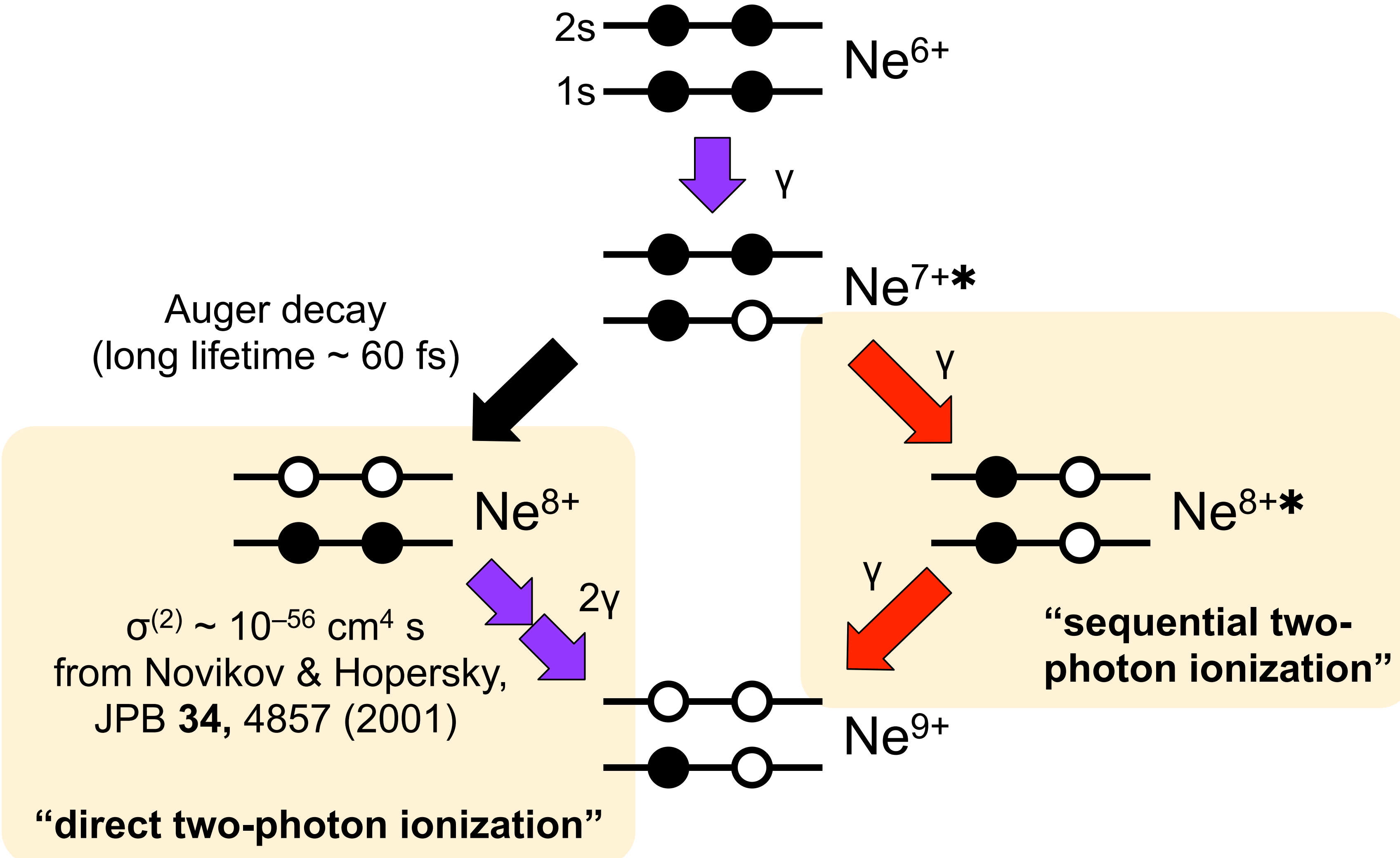
Intensity-dependence of Ne^{9+} / Ne^{8+} ratio

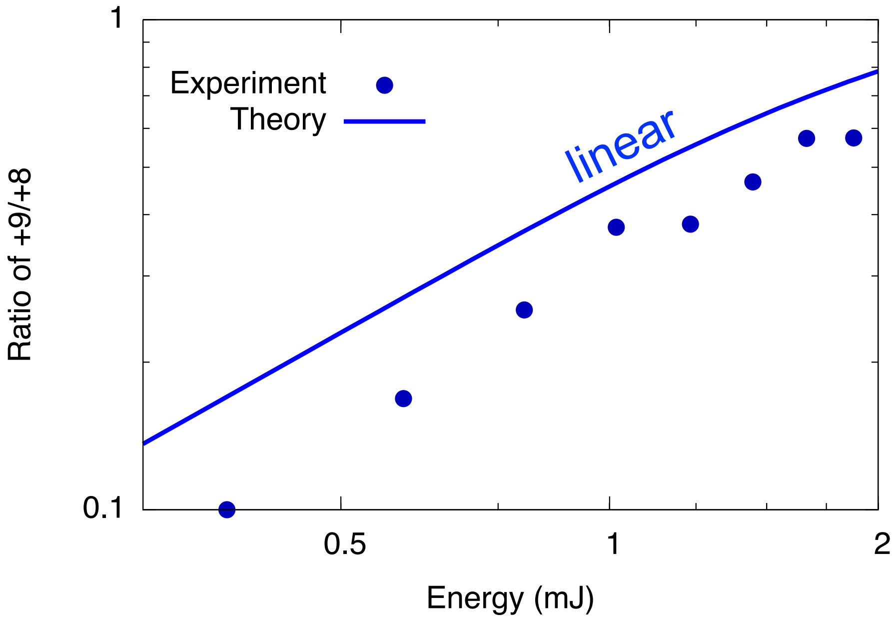


Two-photon ion. mechanisms @1110eV



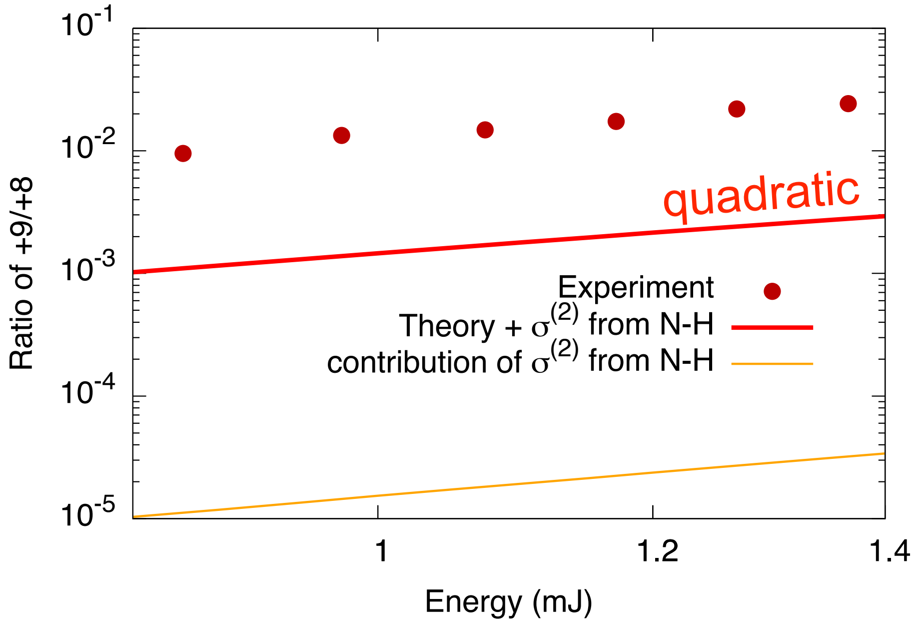
Experiment vs. Theory

1225 eV: above threshold



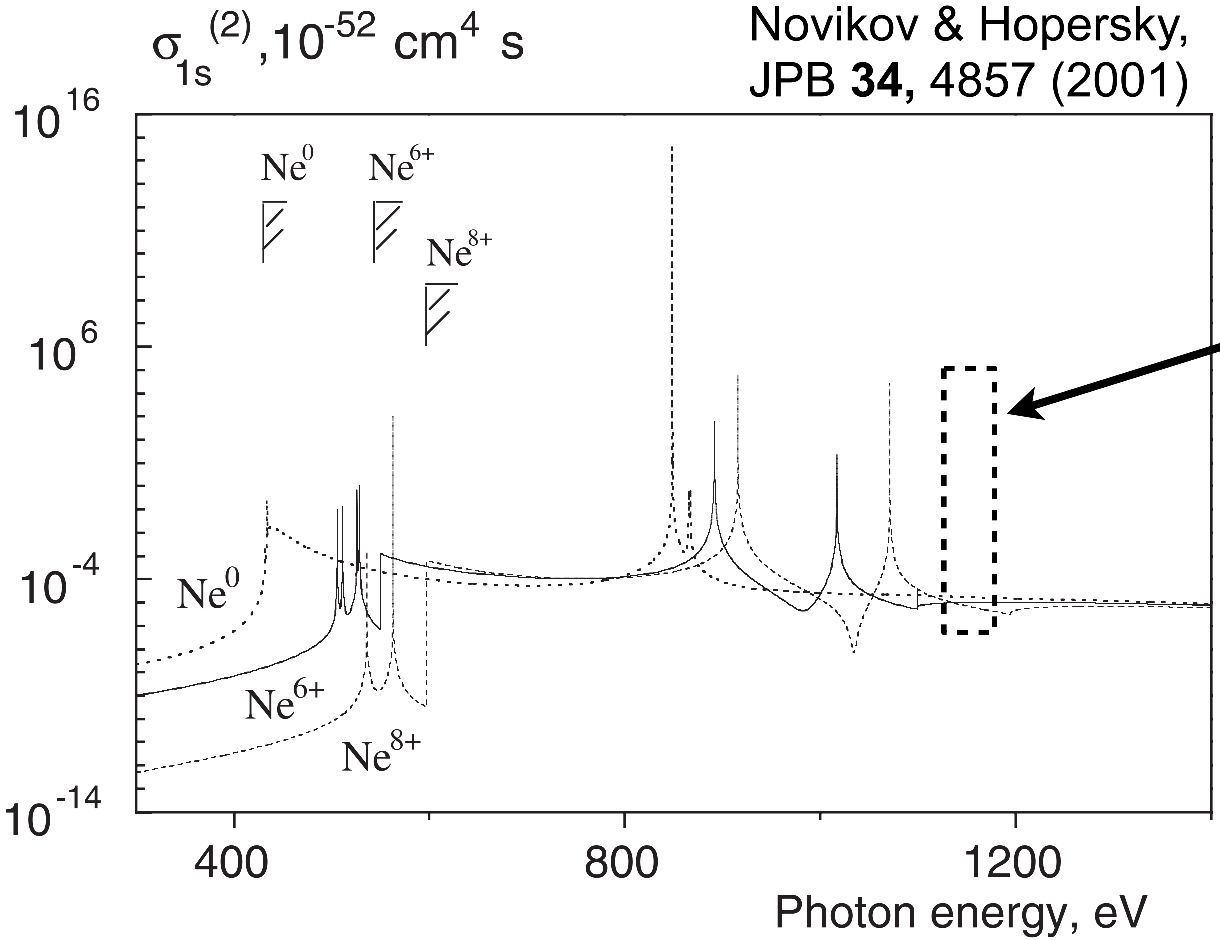
The resonant excitation, Ne^{7+*} $1s^1 2s^2 \rightarrow 1s^0 2s^2 5p^1$ (~1224 eV), is not included in the model.

1110 eV: below threshold



- sequential two-photon ionization: quadratic dependence
- direct two-photon ionization: $\sigma^{(2)}$ is too small

Direct two-photon ionization cross section

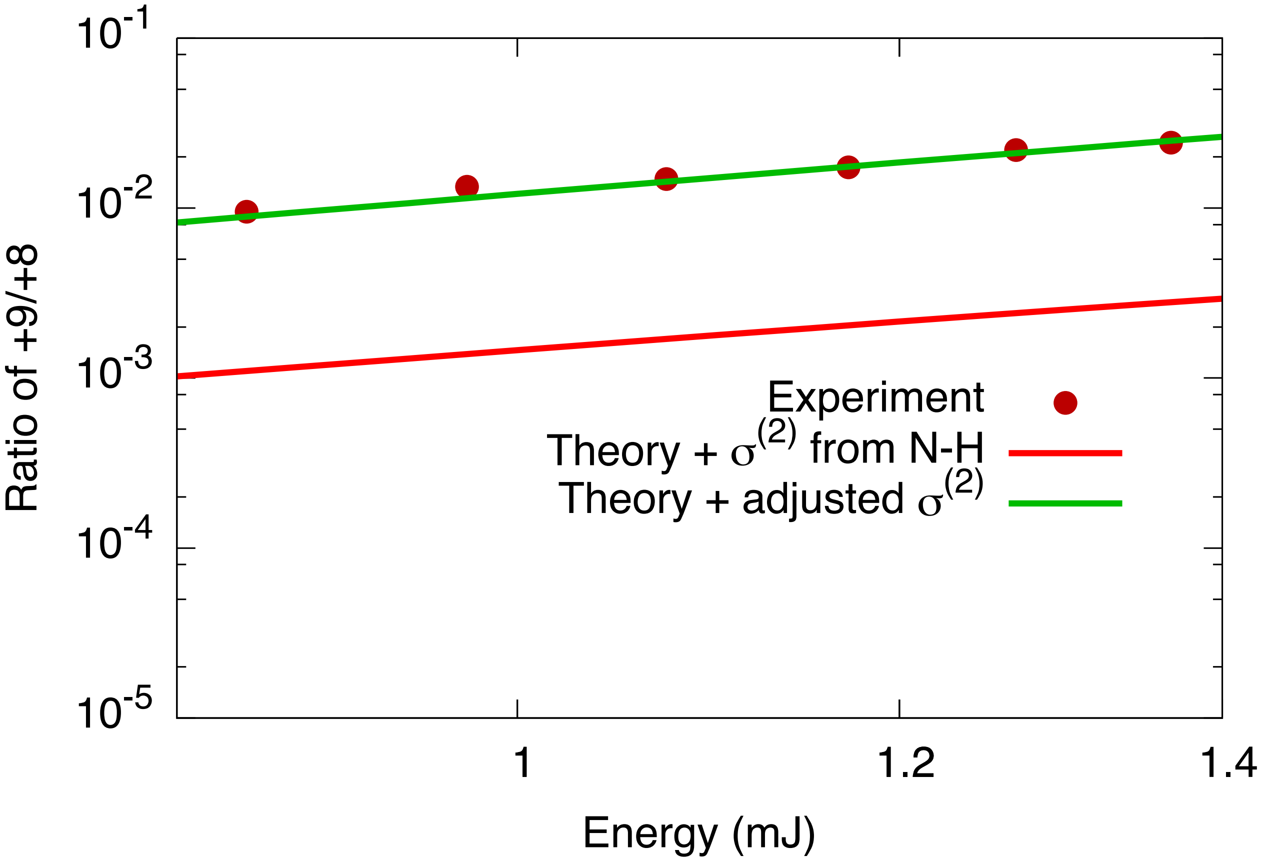


Novikov & Hopersky, JPB **34,** 4857 (2001)

The resonant excitation, Ne^{8+} $1s^2 \rightarrow 1s^1 4p^1$ (~1127 eV), is not included in their calculations.

$\sigma^{(2)}$ can be resonantly enhanced at 1110 eV.

Experiment vs. Theory with adjusted σ$^{(2)}$



1110 eV
below threshold

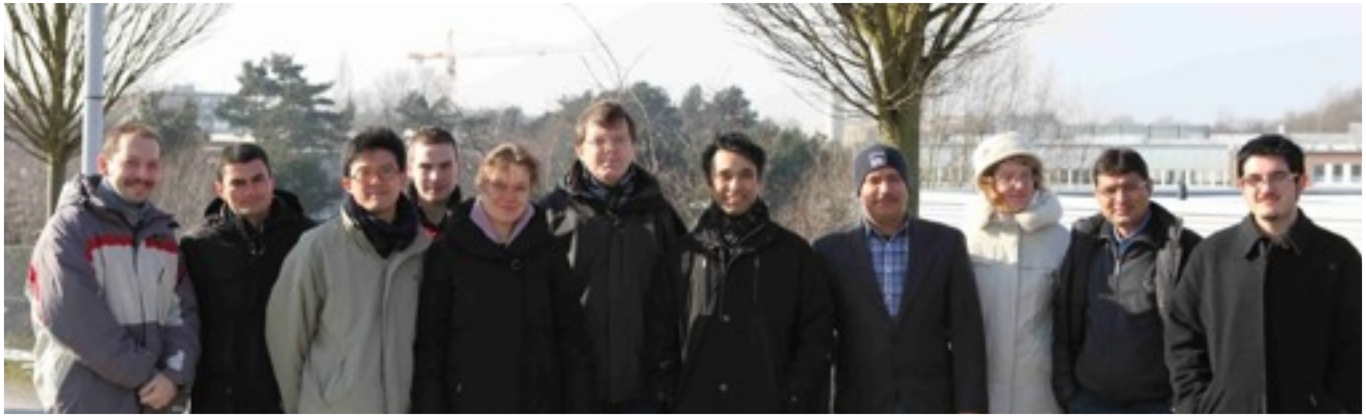
adjusted σ$^{(2)}$ ~ 700 times larger than that from Novikov & Hopersky

Conclusion

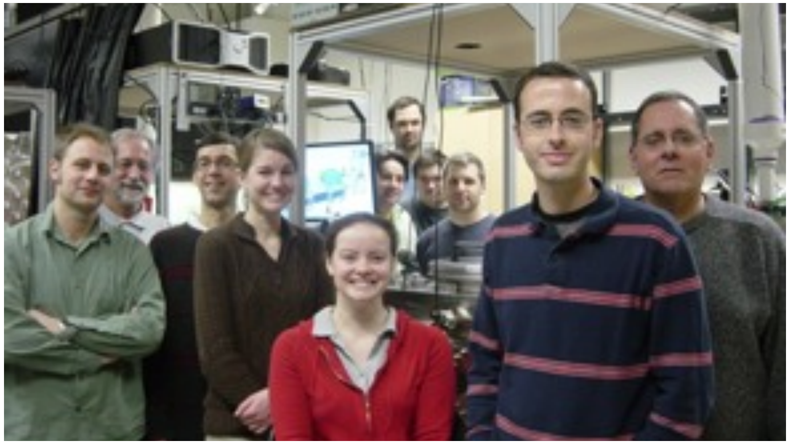
- First experimental evidence of nonlinear absorption in the x-ray regime
- Quadratic dependence of the Ne^{9+} production when the photon energy is below the K-shell threshold of Ne^{8+}
- Nonlinear response from two channels: direct two-photon ionization and sequential two-photon ionization with transient excited states competing with the Auger decay clock
- The direct two-photon ionization cross section is 2–3 orders of magnitude higher than expected from previous calculations.

Acknowledgment

- CFEL Theory Group at DESY



- Agostini – DiMauro group at OSU



conducted at LCLS at the SLAC National Accelerator Lab.

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