Nonlinear x-ray absorption

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HASYLAB Users' Meeting
European XFEL Users' Meeting
January 27, 2011

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Acknowledgments

- Closely related, practically important questions:

  - Does multiphoton absorption play a role in the x-ray regime?

  - Is multiphoton absorption in the x-ray regime primarily sequential or nonsequential?

  - Is sequential multiphoton absorption nonlinear, or is nonlinearity a privilege of nonsequential multiphoton absorption?

  - Does the fact that SASE pulses consist of a series of intense subspikes enhance x-ray FEL-induced damage in materials?

Computational tool: coupled rate equations with ab initio photoionization cross sections and decay rates

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

$$\sigma_{\mathrm{P}}(i,\omega) = \frac{4}{3}\alpha\pi^2\omega N_i \sum_{l_j=|l_i-1|}^{l_i+1} \frac{l_>}{2l_i+1}\left|\int_0^\infty P_{n_i l_i}(r)P_{\varepsilon l_j}(r)\ r\ dr\right|^2$$

$$\Gamma_{\mathrm{A}}(i,jj') = \pi\frac{N_i^{\mathrm{H}}N_{jj'}}{2l_i+1}\sum_{L=|l_j-l_{j'}|}^{l_j+l_{j'}}\sum_{S=0}^{1}\sum_{l_{i'}}(2L+1)(2S+1)|M_{LS}(j,j',i,i')|^2$$

$$R_K(j,j',i,i') = \int_0^\infty\int_0^\infty P_{n_j l_j}(r_1)P_{n_{j'}l_{j'}}(r_2)\frac{r_<^K}{r_>^{K+1}}P_{n_i l_i}(r_1)P_{\varepsilon l_{i'}}(r_2)\ dr_1\ dr_2$$

$$\Gamma_{\mathrm{F}}(i,j) = \frac{4}{3}\alpha^3(I_i-I_j)^3\frac{N_i^{\mathrm{H}}N_j}{4l_j+2}\cdot\frac{l_>}{2l_i+1}\left|\int_0^\infty P_{n_i l_i}(r)P_{n_j l_j}(r)\ r\ dr\right|^2$$
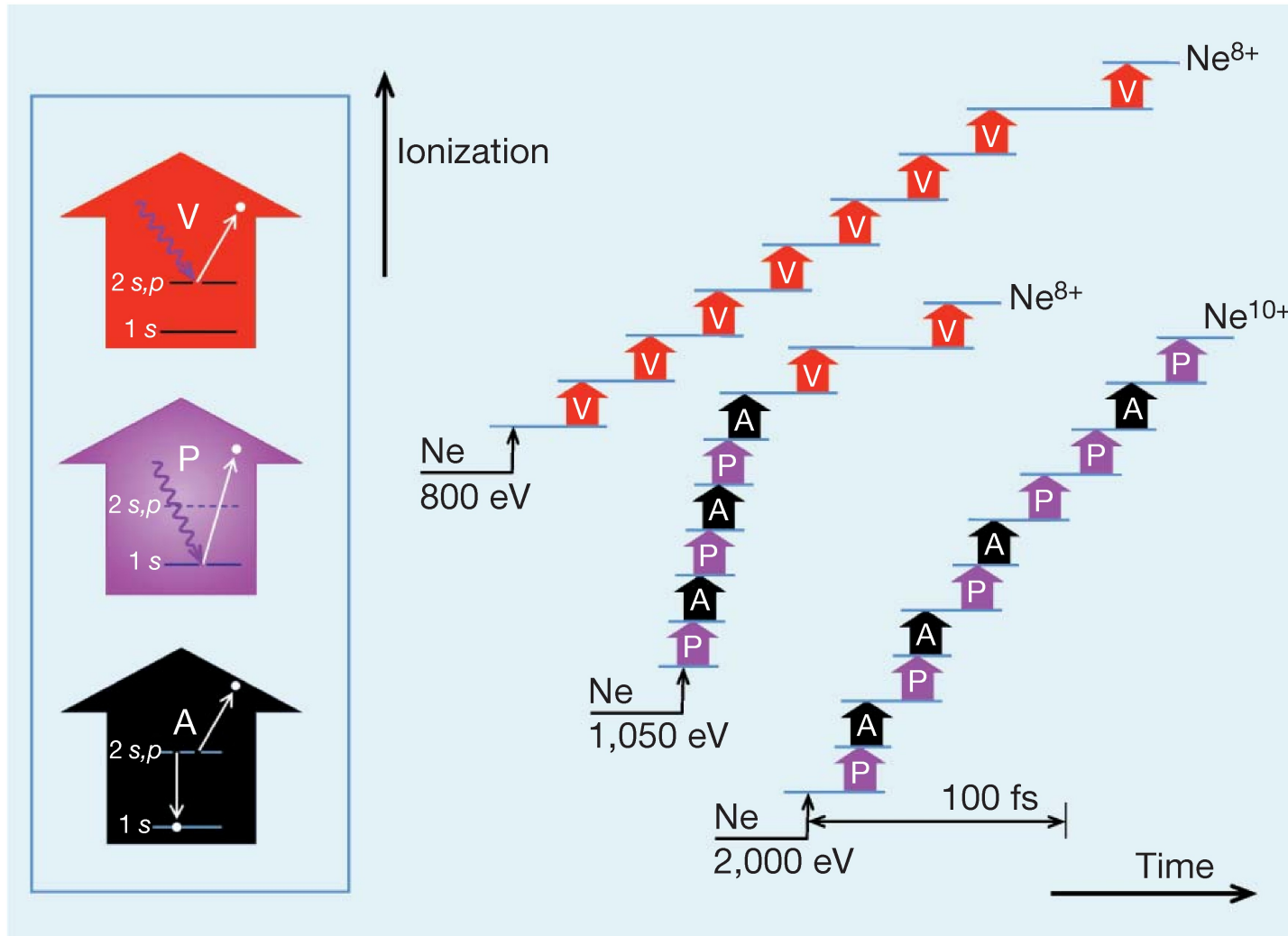
XATOM toolkit, implemented by Sang-Kil Son

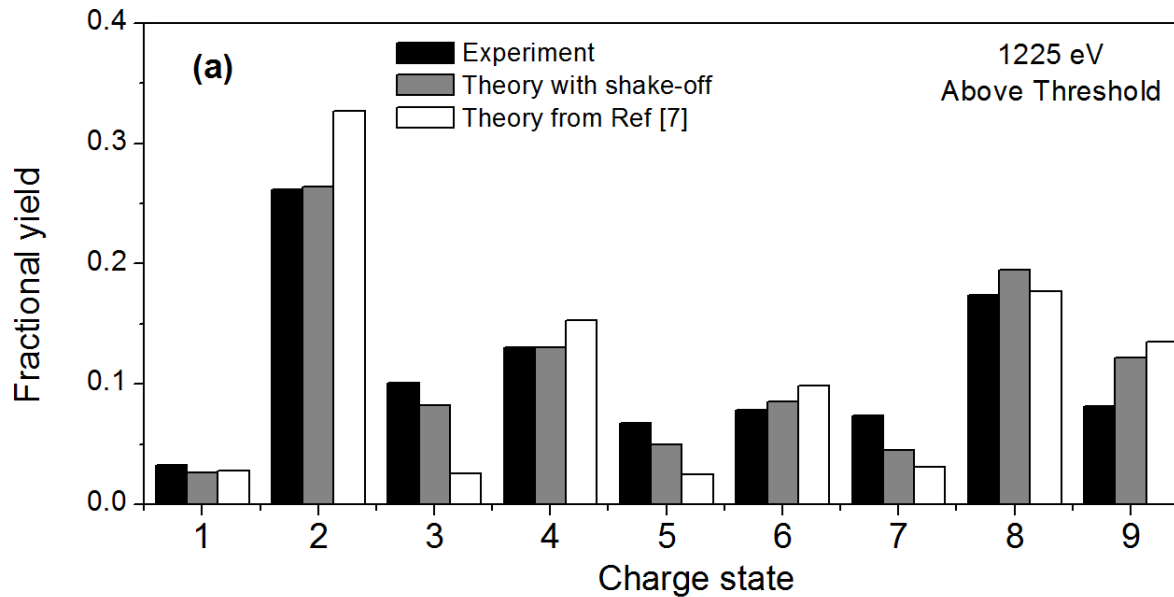[S.-K. Son, L. Young, and R. Santra, Phys. Rev. A, in press (2011)]

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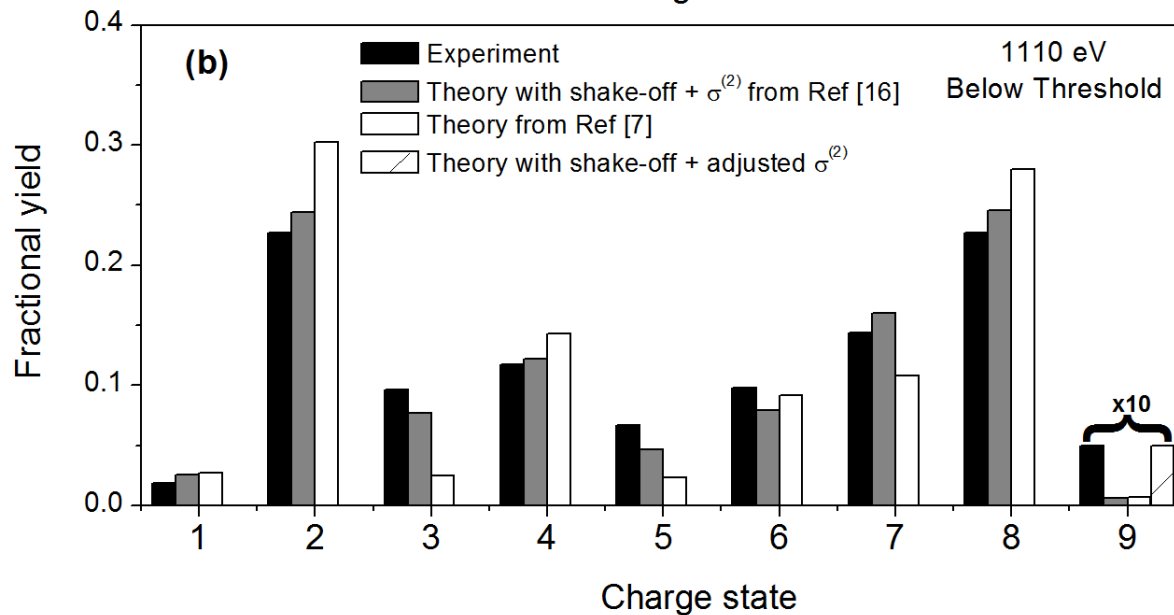# Sequential multiphoton ionization of neon

L. Young *et al.*, Nature **466**, 56 (2010)
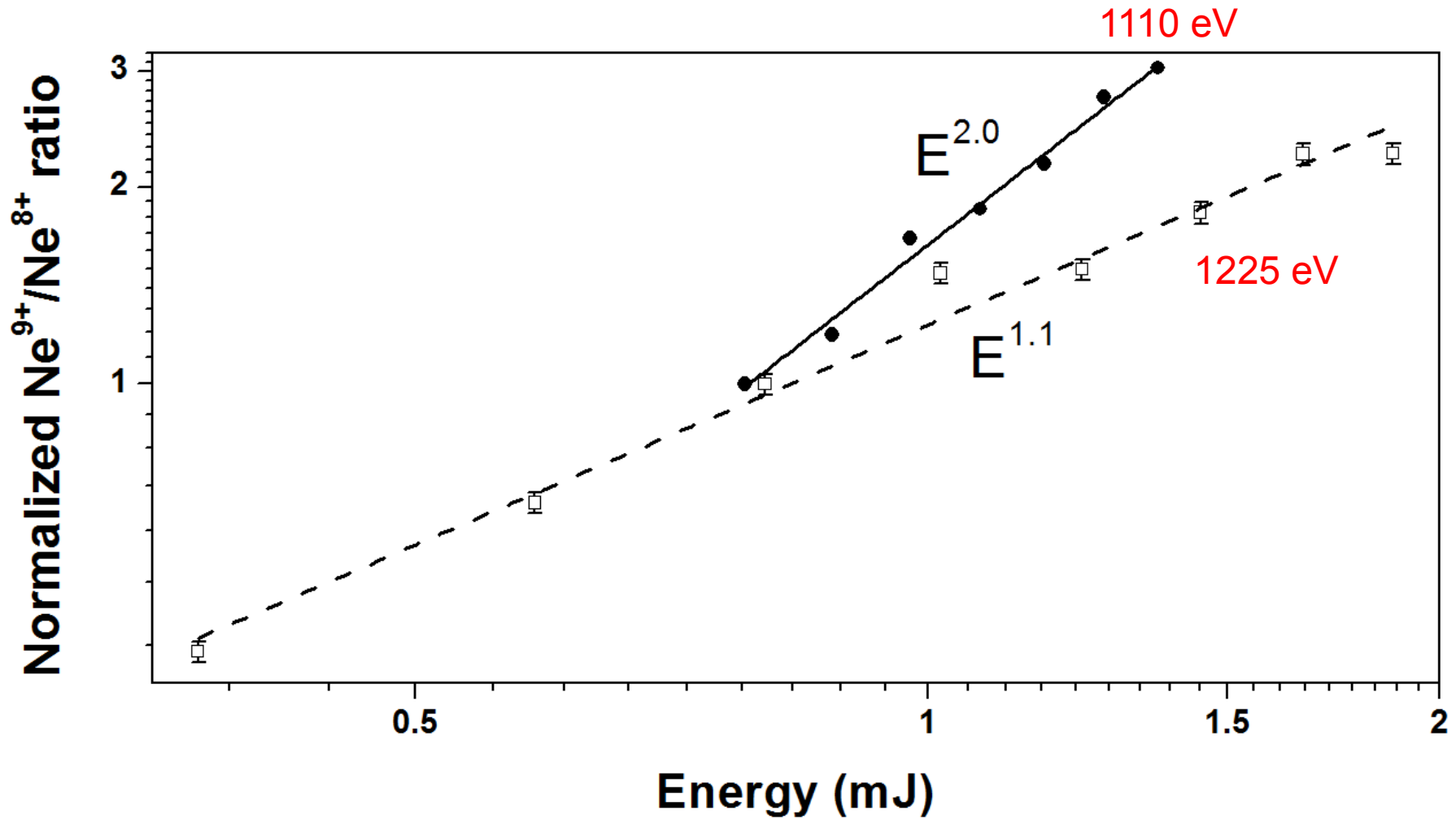
Neon charge-state distributions

Comparison of experiment with Sang-Kil Son's calculations
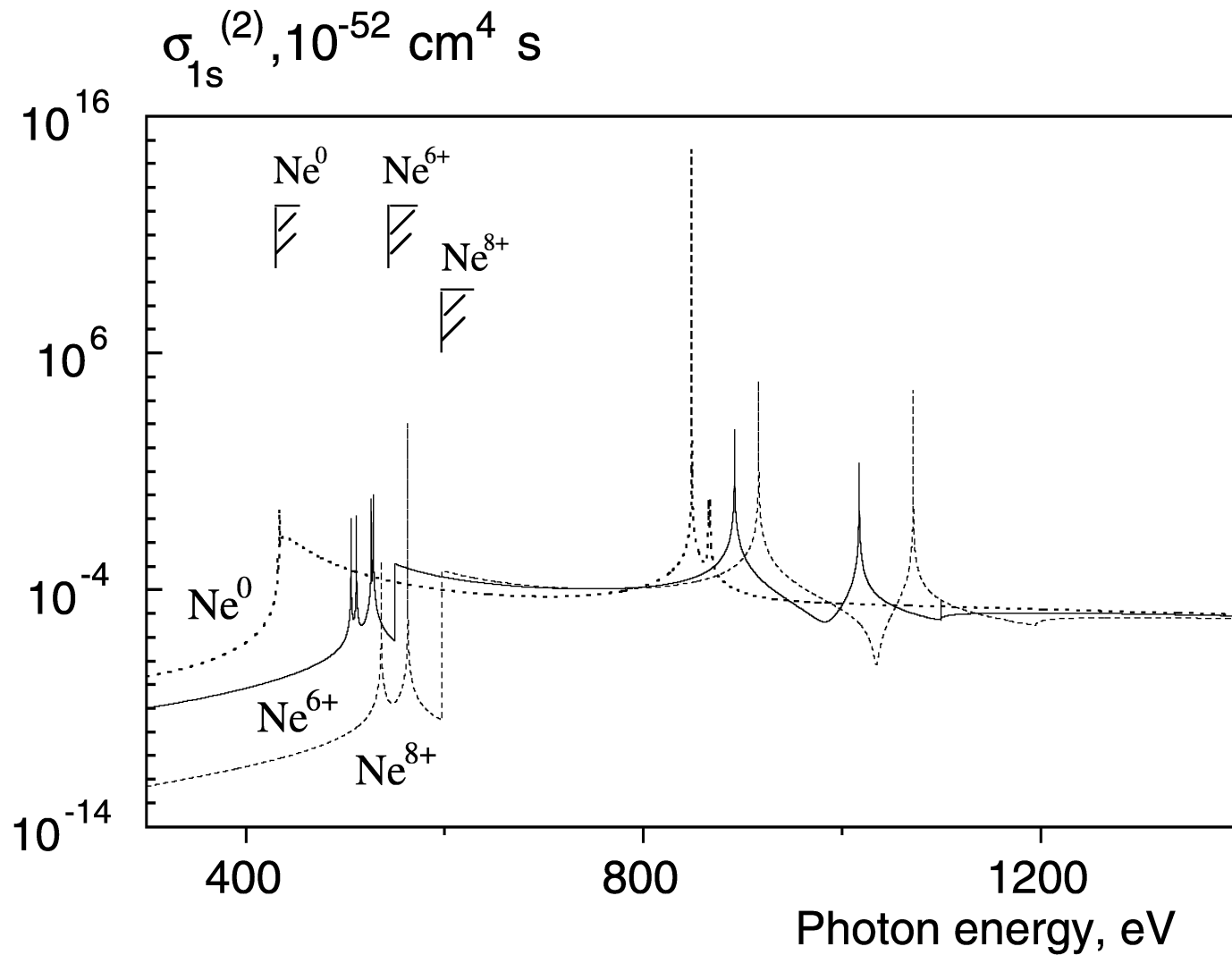
G. Doumy *et al.*, submitted

Nonlinear production of Ne^{9+}: observation

G. Doumy *et al.*, submitted
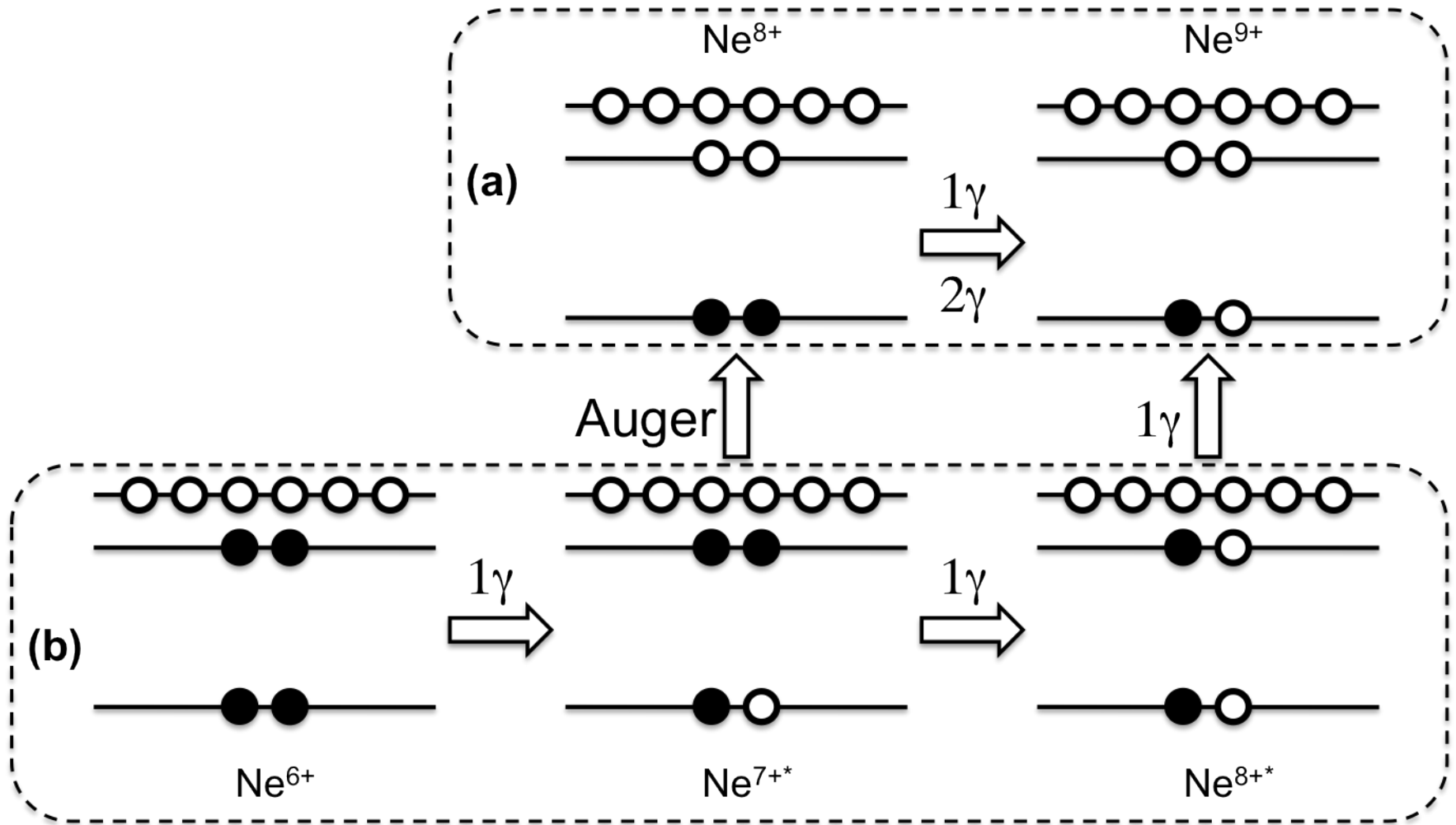
Two-photon ionization cross section of Ne^{8+}

$\sigma_{1s}^{(2)}, 10^{-52}\ cm^4\ s$

S. A. Novikov and
A. N. Hopersky,
J. Phys. B **34**,
4857 (2001)

G. Doumy *et al.*, submitted
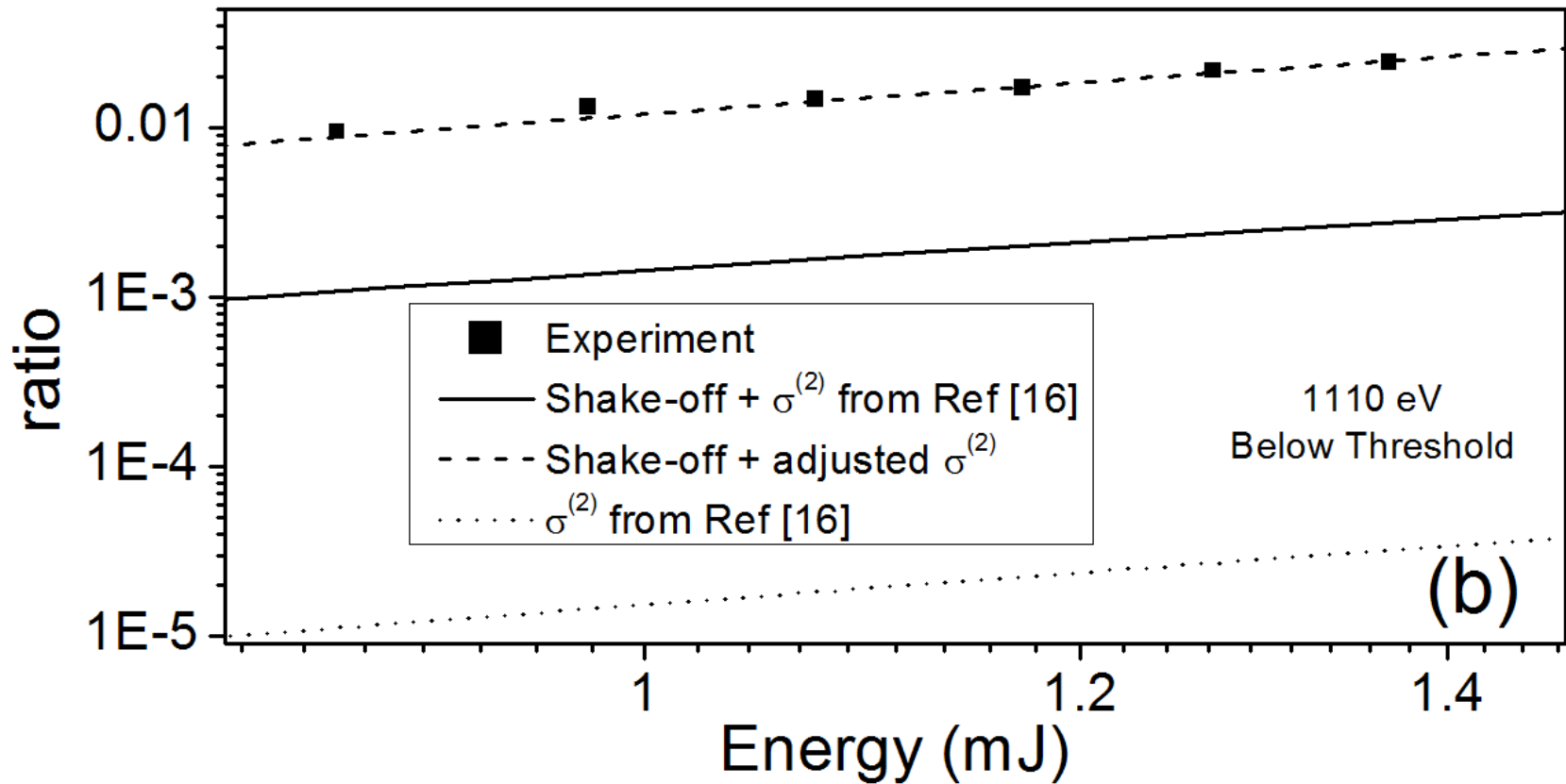
G. Doumy *et al.*, submitted

Impact of chaoticity of SASE radiation
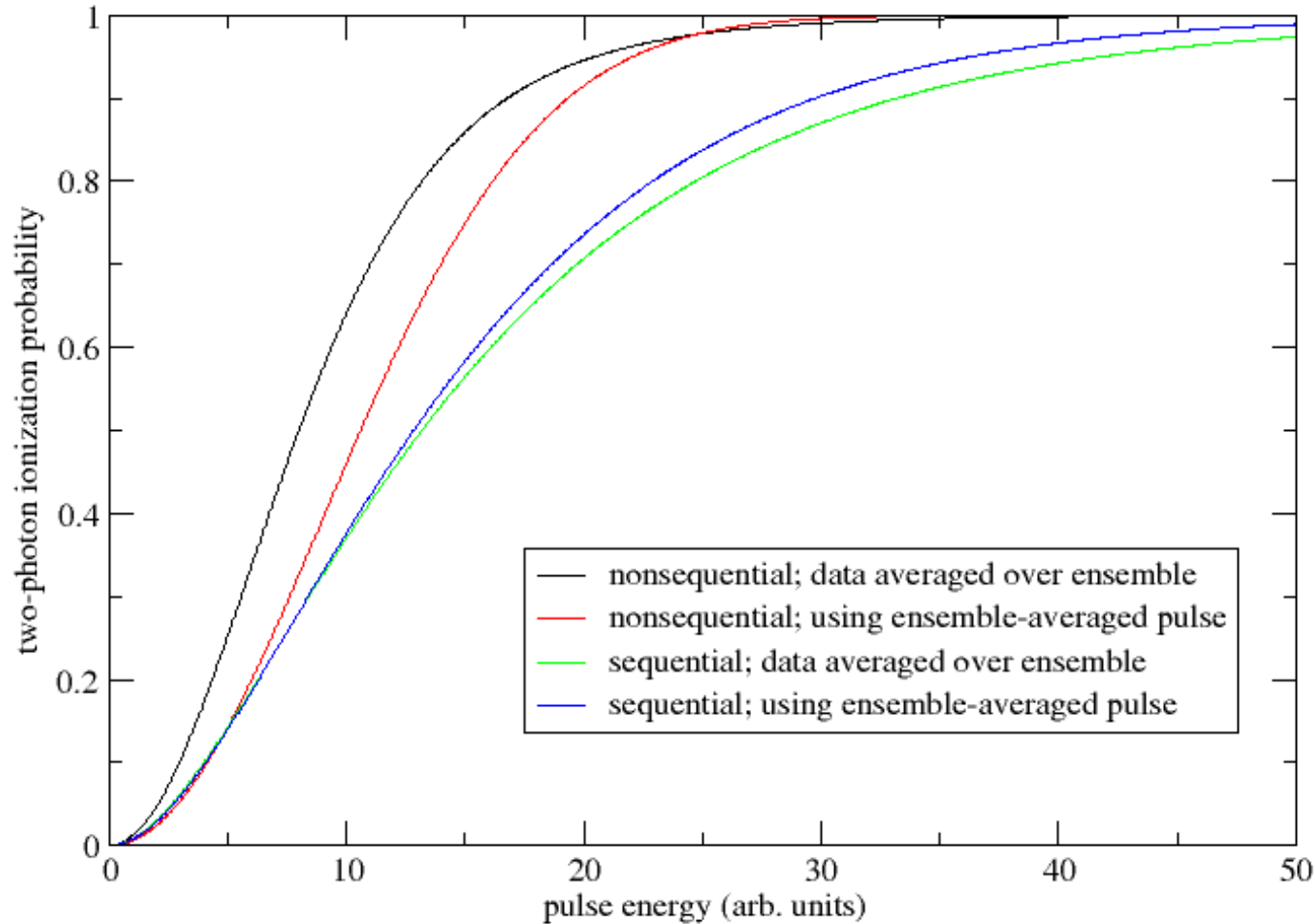
- Sequential vs. nonsequential two-photon ionization

- See also N. Rohringer and R. Santra, Phys. Rev. A **76**, 033416 (2007)

R. Santra, unpublished

Conclusions

> Multiphoton absorption is important for experiments using intense x-ray FEL radiation.

> Multiphoton absorption in the x-ray regime is predominantly sequential.

> Sequential multiphoton absorption can display nonlinearities.

> Multiphoton absorption in the x-ray regime is quite insensitive to the spiky pulse structure of SASE radiation.

> There is first evidence for a nonsequential process in the x-ray regime.