AC-Stark splitting of Auger spectra under intense x-ray radiation

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Overview

Neon under intense 908 eV radiation

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Density matrix equations for the double continuum

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- Results on Auger kinetic energy spectrum and ionization yields

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- Density matrix equations for the double continuum
- Results on Auger kinetic energy spectrum and ionization yields
- The experimental aspect

Ionization scheme

Some history

Rabi flopping of the Auger spectra has been studied in Ne$(1s^{-1} - 3p)$ excitation (Resonant Auger State)

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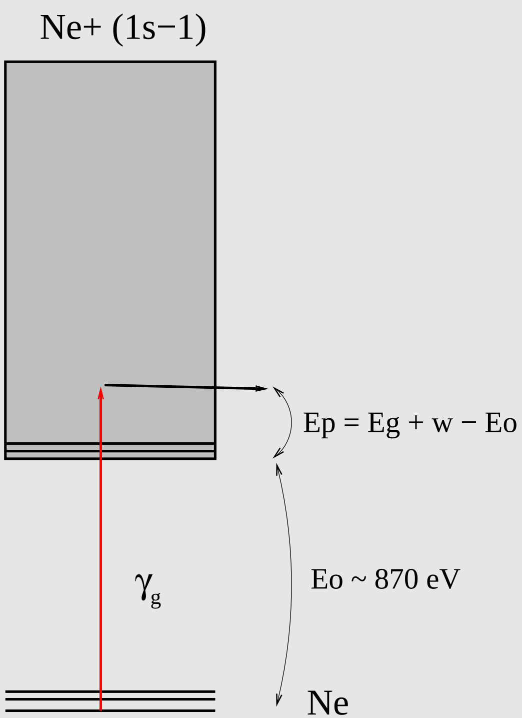
Can be done something similar to normal Auger line and how?

Neon under intense x-ray radiation

Photoionization step

$$\text{Ne}(1s^2 2s^2 2p^6) \rightarrow \text{Ne}^+(1s^1 2s^2 2p^6) + e_p^-$$



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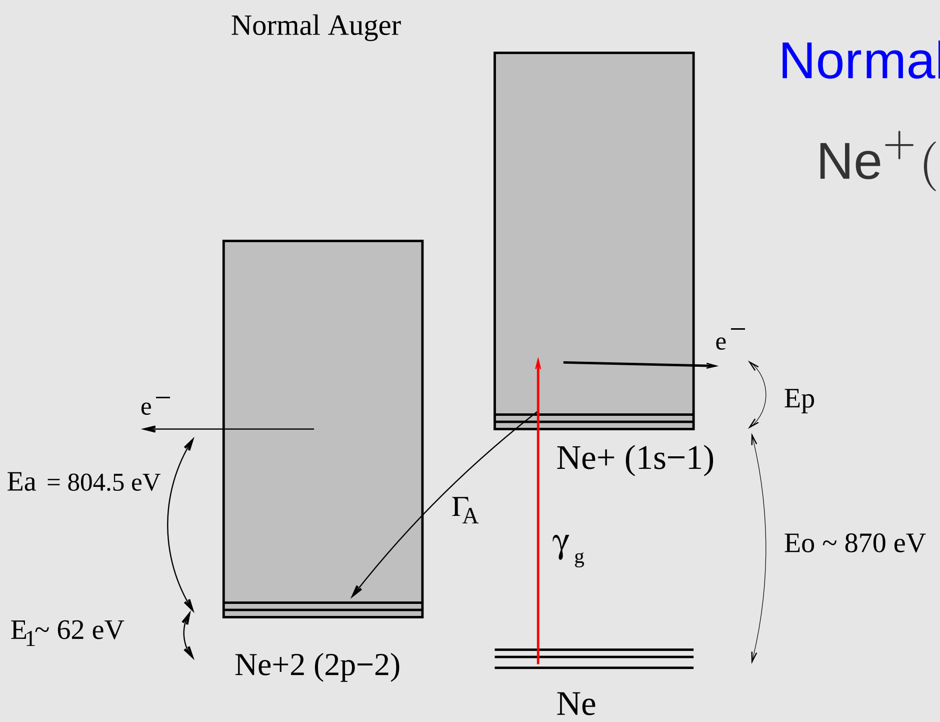

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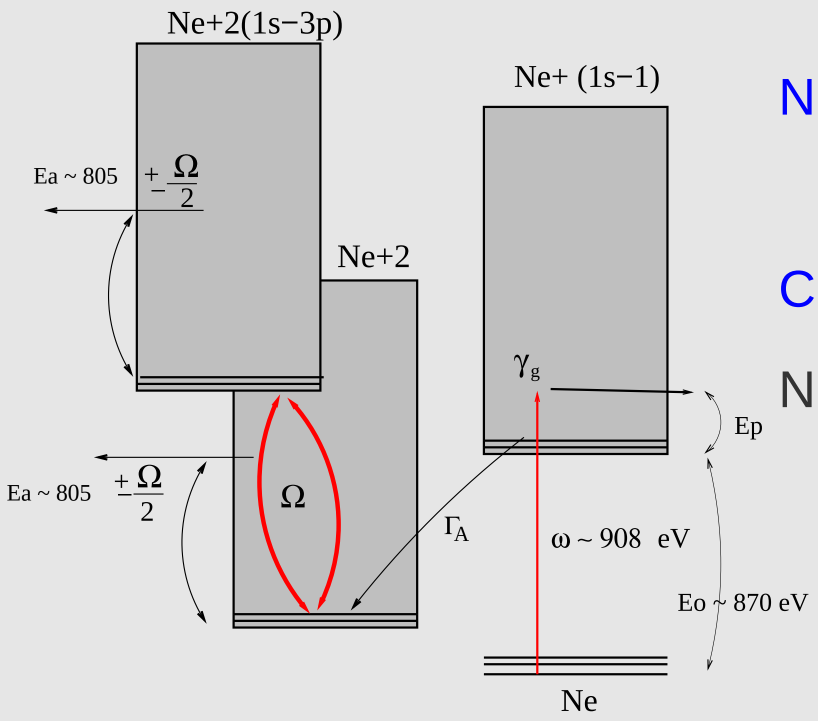

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Core-resonant Auger transition

$$\begin{aligned} \text{Ne}^+(1s^1 2s^2 2p^6) &\rightarrow \text{Ne}^{+2}(1s^2 2s^2 2p^4) + e_a \\ &\leftrightarrow \text{Ne}^{+2}(1s^1 2s^2 2p^4)(3p)) + e_a^- \end{aligned}$$

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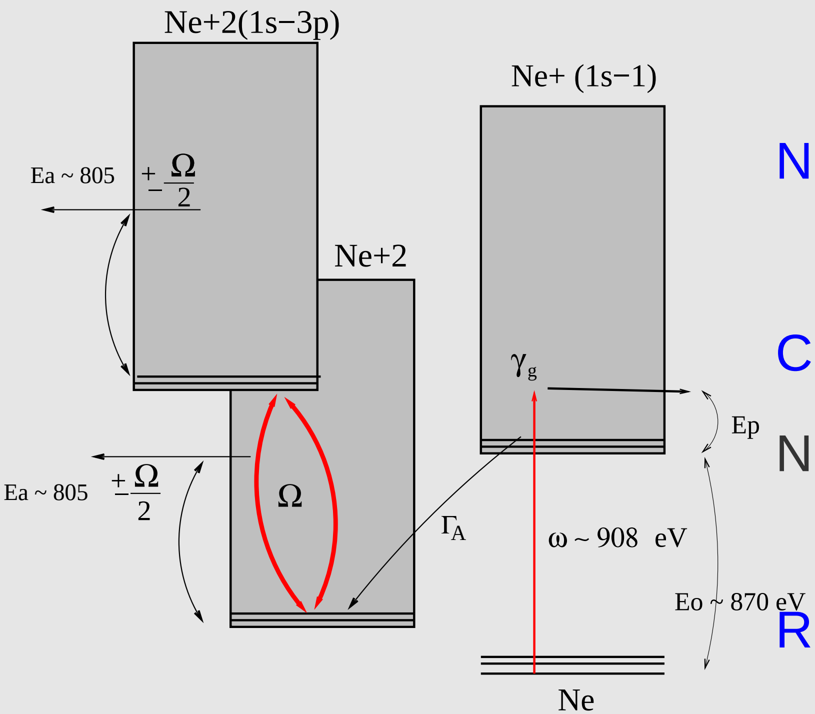

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Rabi K-shell excitation 1s-3p

$$\text{Ne}^{+2}(1s^2 2s^2 2p^4) + e_a \rightarrow \text{Ne}^{+2}(1s^1 2s^2 2p^4)\, 3p$$

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Excited Ne$^{+2}(1s^{-1} - 3p)$ states

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$\|a'\rangle$	$E_{a'}$ (eV)	C=Ne$^{+2}(1s^1 2s^2 2p^4, {}^2L)$
1	907.75	$[C]^2D(3p^1)^1P_1$
2	907.90	$[C]^2P(3p^1)^3P_1$
3	908.06	$[C]^2D(3p^1)^1F_3$
4	908.48	$[C]^2P(3p^1)^3D_3$
5	908.51	$[C]^2D(3p^1)^3D_2$
6	908.49	$[C]^2P(3p^1)^1D_2$
7	908.78	$[C]^2D(3p^1)^1D_2$

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$\|a'\rangle$	$E_{a'}$ (eV)	Ne$^{+2}(1s^1 2s^2)$	$gf_{aa'}(\times 10^{-2})$
1	907.75	$(2p^4, {}^1D)^2D(3p^1)^1P_1$	2.3338
2	907.90	$(2p^4, {}^3P)^2P(3p^1)^3P_1$	0.20991
3	908.06	$(2p^4, {}^1D)^2D(3p^1)^1F_3$	8.1881
4	908.48	$(2p^4, {}^3P)^2P(3p^1)^3D_3$	0.13141
5	908.51	$(2p^4, {}^1D)^2D(3p^1)^3D_2$	0.23322
6	908.49	$(2p^4, {}^3P)^2P(3p^1)^1D_2$	4.4888
7	908.78	$(2p^4, {}^1D)^2D(3p^1)^1D_2$	1.2714

The density matrix equations for the double continuum

The atomic states involved

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- Coupled Ne^{+2} states (ground and excited)

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- The (dissipative) environment for Ne^{+}

$$\text{Ne}^{+} \longrightarrow |R\rangle = Ne^{+1}(1s^2 2s^2 2p^5) + \omega_r, \quad \textit{Fluorescence}$$

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- The dissipative environment for Ne^{+2}

$$\begin{aligned} \text{Ne}^{+2}(1s^{-1} - 3p) &\longrightarrow |F_1\rangle = Ne^{+3}(1s^2 2s^2 2p^3), \text{ Auger decay} \\ &\longrightarrow |F_2\rangle = Ne^{+3}(1s^1 2s^2 2p^4) \quad \text{photoionization} \end{aligned}$$

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Full Density Matrix Equations (28)

$$
\begin{aligned}
\dot{\rho}_{gg}(t) &= 2Im \sum_I D_{GI}\rho_{IG}, \\
\dot{\rho}_{ii}(\mathbf{k}_i, t) &= 2Im\left[D_{IG}\rho_{GI}\right] + 2Im \sum_A V_{IA}\rho_{AI} \\
&+ 2Im \sum_R D_{IR}\rho_{RI} \\
\dot{\rho}_{aa}(\mathbf{k}_a, \mathbf{k}_i, t) &= 2Im\left[V_{AI}\rho_{IA}\right] + 2Im\left[D_{AA'}\rho_{A'A}\right] \\
\dot{\rho}_{a'a'}(\mathbf{k}_a, \mathbf{k}_i, t) &= -2Im\left[D_{AA'}\rho_{A'A}\right] + 2Im \sum_{F_1} V_{A'F_1}\rho_{F_1A'} \\
&+ 2Im \sum_{F_2} D_{A'F_2}\rho_{F_2A'} \\
i\dot{\rho}_{aa'}(\mathbf{k}_a, \mathbf{k}_i, t) &= E_{AA'}\rho_{AA'} + D_{AA'}(\rho_{A'A'} - \rho_{AA}) + V_{AI}\rho_{IA'} \\
&- \sum_{F_1} \rho_{AF_1}V_{F_1A'} - \sum_{F_2} \rho_{AF_2}D_{F_2A'} \\
i\dot{\rho}_{GI}(t) &= \ldots \\
\ldots &\ldots \ldots
\end{aligned}
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- Rotating wave approximation (RWA)
- Elimination of the continuum
- Keeping terms up to the first order to the electric field (not restrictive) ($I < 6 \times 10^{18}$ W/cm^2)
- no interaction between Auger and photo-electrons are allowed (far from ionization thresholds)

DME in terms of $\gamma_g(t), \gamma_{a'}(t), \Gamma_i, \Gamma_{a'}$ and Rabi $\Omega_{aa'}(t)$

$$
\begin{aligned}
\dot\sigma_{gg}(t) &= -\gamma_g \sigma_{gg}, \\
\dot\sigma_{ii}(\varepsilon_i, t) &= -\Gamma_i \sigma_{ii} + Im\left[\Omega^\star_{ig}\sigma_{gi}\right], \\
\dot\sigma_{aa}(\varepsilon_i, \varepsilon_a, t) &= -Im\left[\Omega^\star_{a'a}\sigma_{aa'}\right] + 2Im\left[V_{ai}\sigma_{ia}\right], \\
\dot\sigma_{a'a'}(\varepsilon_i, \varepsilon_a, t) &= -\bar\gamma_{a'}\sigma_{a'a'} + Im\left[\Omega^\star_{a'a}\sigma_{aa'}\right], \\
i\dot\sigma_{aa'}(\varepsilon_i, \varepsilon_a, t) &= (E_{aa'} + \omega - i\frac{\bar\gamma_{a'}}{2})\sigma_{aa'} + \frac{\Omega_{aa'}}{2}(\sigma_{a'a'} - \sigma_{aa}) + V_{ai}\sigma_{ia'}, \\
i\dot\sigma_{gi}(\varepsilon_i, t) &= (E_{gi} + \omega - i\frac{\gamma_g + \Gamma_i}{2})\sigma_{gi} - \frac{1}{2}\Omega_{gi}\ \sigma_{gg} \\
i\dot\sigma_{ia}(\varepsilon_i, \varepsilon_a, t) &= (E_{ia} - i\frac{\Gamma_i}{2})\sigma_{ia} + \frac{1}{2}\Omega^*_{ig}\sigma_{ga} - \frac{1}{2}\Omega^*_{a'a}\sigma_{ia'} - V_{ia}\ \sigma_{ii} \\
i\dot\sigma_{ia'}(\varepsilon_i, \varepsilon_a, t) &= (E_{ia'} + \omega - i\frac{\Gamma_i + \bar\gamma_{a'}}{2})\sigma_{ia'} + \frac{1}{2}\Omega^*_{ig}\sigma_{ga'} - \frac{1}{2}\Omega_{aa'}\sigma_{ia}, \\
i\dot\sigma_{ga}(\varepsilon_i, \varepsilon_a, t) &= (E_{ga} + \omega - i\frac{\gamma_g}{2})\sigma_{ga} - \frac{1}{2}\Omega^*_{a'a}\sigma_{ga'} - V_{ia}\sigma_{gi}, \\
i\dot\sigma_{ga'}(\varepsilon_i, \varepsilon_a, t) &= (E_{ga'} + 2\omega - i\frac{\gamma_g + \bar\gamma_{a'}}{2})\sigma_{ga'} - \frac{1}{2}\Omega_{aa'}\sigma_{ga}.
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\dot{\sigma}_{ii}(\varepsilon_i, t) &= -\Gamma_i \sigma_{ii} + Im\left[\Omega^{\star}_{ig}\sigma_{gi}\right], \\
\dot{\sigma}_{aa}(\varepsilon_i, \varepsilon_a, t) &= -Im\left[\Omega^{\star}_{a'a}\sigma_{aa'}\right] + 2Im\left[V_{ai}\sigma_{ia}\right], \\
\dot{\sigma}_{a'a'}(\varepsilon_i, \varepsilon_a, t) &= -\bar{\gamma}_{a'}\sigma_{a'a'} + Im\left[\Omega^{\star}_{a'a}\sigma_{aa'}\right], \\
i\dot{\sigma}_{aa'}(\varepsilon_i, \varepsilon_a, t) &= (E_{aa'} + \omega - i\frac{\bar{\gamma}_{a'}}{2})\sigma_{aa'} + \frac{\Omega_{aa'}}{2}(\sigma_{a'a'} - \sigma_{aa}) + V_{ai}\sigma_{ia'}, \\
i\dot{\sigma}_{gi}(\varepsilon_i, t) &= (E_{gi} + \omega - i\frac{\gamma_g + \Gamma_i}{2})\sigma_{gi} - \frac{1}{2}\Omega_{gi}\sigma_{gg} \\
i\dot{\sigma}_{ia}(\varepsilon_i, \varepsilon_a, t) &= (E_{ia} - i\frac{\Gamma_i}{2})\sigma_{ia} + \frac{1}{2}\Omega^{*}_{ig}\sigma_{ga} - \frac{1}{2}\Omega^{*}_{a'a}\sigma_{ia'} - V_{ia}\sigma_{ii} \\
i\dot{\sigma}_{ia'}(\varepsilon_i, \varepsilon_a, t) &= (E_{ia'} + \omega - i\frac{\Gamma_i + \bar{\gamma}_{a'}}{2})\sigma_{ia'} + \frac{1}{2}\Omega^{*}_{ig}\sigma_{ga'} - \frac{1}{2}\Omega_{aa'}\sigma_{ia}, \\
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i\dot{\sigma}_{ga'}(\varepsilon_i, \varepsilon_a, t) &= (E_{ga'} + 2\omega - i\frac{\gamma_g + \bar{\gamma}_{a'}}{2})\sigma_{ga'} - \frac{1}{2}\Omega_{aa'}\sigma_{ga}.
\end{aligned}$$

Coarse-grained DME for the Auger-electron

$$
\begin{aligned}
\dot{\sigma}_{gg} &= -\gamma_g \sigma_{gg}, \\
\dot{\sigma}_{ii}(\varepsilon_a, t) &= -\Gamma_i \sigma_{ii} + \gamma_g \sigma_{gg}, \\
\dot{\sigma}_{aa}(\varepsilon_a, t) &= -Im\left[\Omega^{\star}_{a'} \sigma_{aa'}\right] + Im\left[\bar{\Delta}(\Omega^{+}_{a'} - \Omega^{-}_{a'})\right] \sigma_{ii}, \\
\dot{\sigma}_{a'a'}(\varepsilon_a, t) &= -\Gamma_{a'} \sigma_{a'a'} + Im\left[\Omega^{\star}_{a'} \sigma_{aa'}\right], \\
i\dot{\sigma}_{aa'}(\varepsilon_a, t) &= \bar{\delta} \sigma_{aa'} - \frac{\Omega_{a'}}{2}(\sigma_{a'a'} - \sigma_{aa}) + \frac{\Omega_{a'}}{4}(\Omega^{+}_{a'} - \Omega^{-}_{a'}) \sigma_{ii}.
\end{aligned}
$$

Integrated over the energies of the photoionized electron ($\mathbf{k}_i$).

All derivatives of the coherences that include the photoelectron state were set to zero (adiabatic approximation) .

$$
S(\varepsilon_a) = \int_{-\infty}^{+\infty} dt \left[\dot{\sigma}_{aa}(\varepsilon_a, t) + \dot{\sigma}_{a'a'}(\varepsilon_a, t)\right]
$$

$$
P_a(t) = \int d\varepsilon_a \sigma_{aa}(\varepsilon_a, t), \quad P_{a'}(t) = \int d\varepsilon \sigma_{a'a'}(\varepsilon_a, t),
$$

Auger spectrum analytical formula

For long pulses

$$S(\varepsilon_a) = \frac{\Gamma_{ia}}{4\pi}[\frac{1-\delta_{a'}/\bar{\Omega}_{a'}}{(\varepsilon_a - \varepsilon_a^{(0)} - \frac{\delta_{a'}-\bar{\Omega}_{a'}}{2})^2 + \frac{\Gamma_i^2}{4}} + \frac{1+\delta_{a'}/\bar{\Omega}_{a'}}{(\varepsilon_a - \varepsilon_a^{(0)} - \frac{\delta_{a'}+\bar{\Omega}_{a'}}{2})^2 + \frac{\Gamma_i^2}{4}}].$$

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$\varepsilon_a^{(0)}$ Normal Auger energy

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- As a rule of thumb, the following ratios matter:

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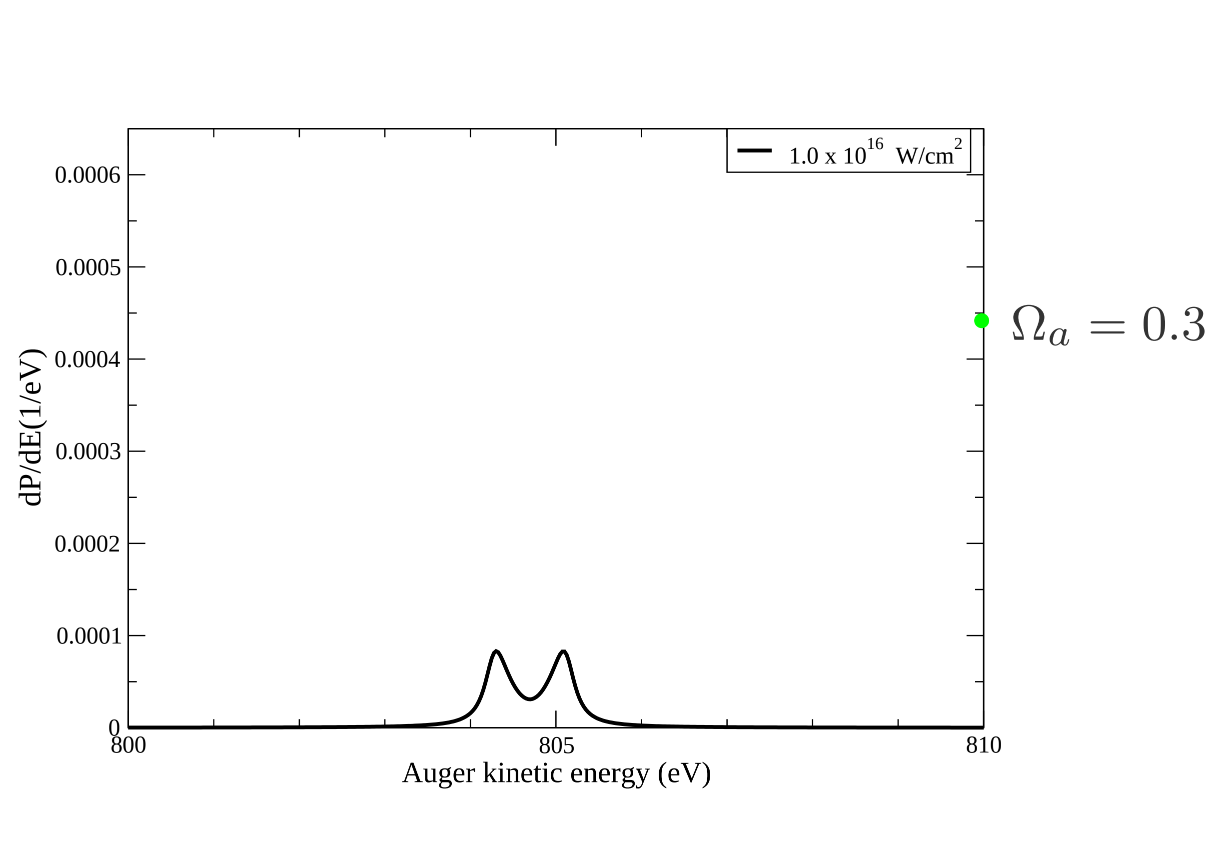
Results for Neon under 48.8 fs pulse

Core resonant Auger kinetic spectra



- $\Omega_a = 0.3 \sim \Gamma_i$

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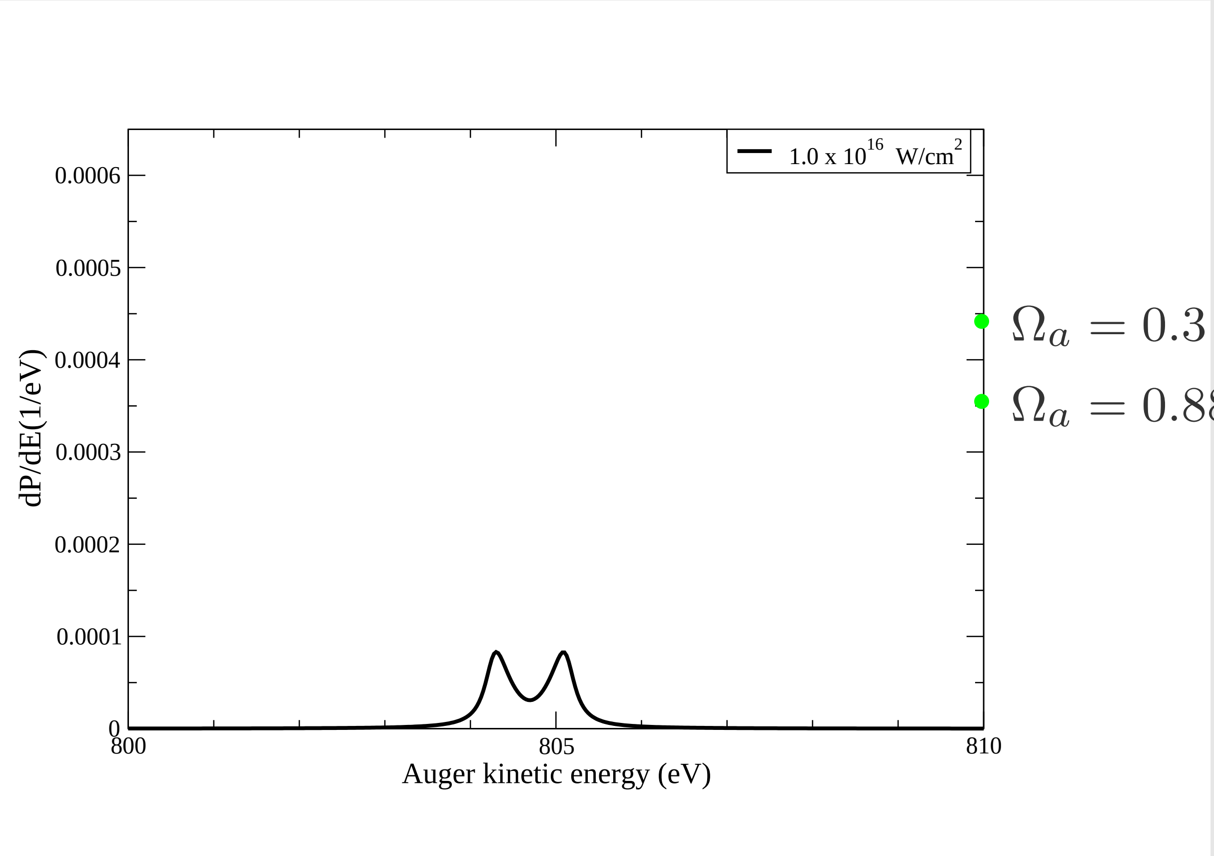

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- $\Omega_a = 0.88 \sim 3\Gamma_i$ eV

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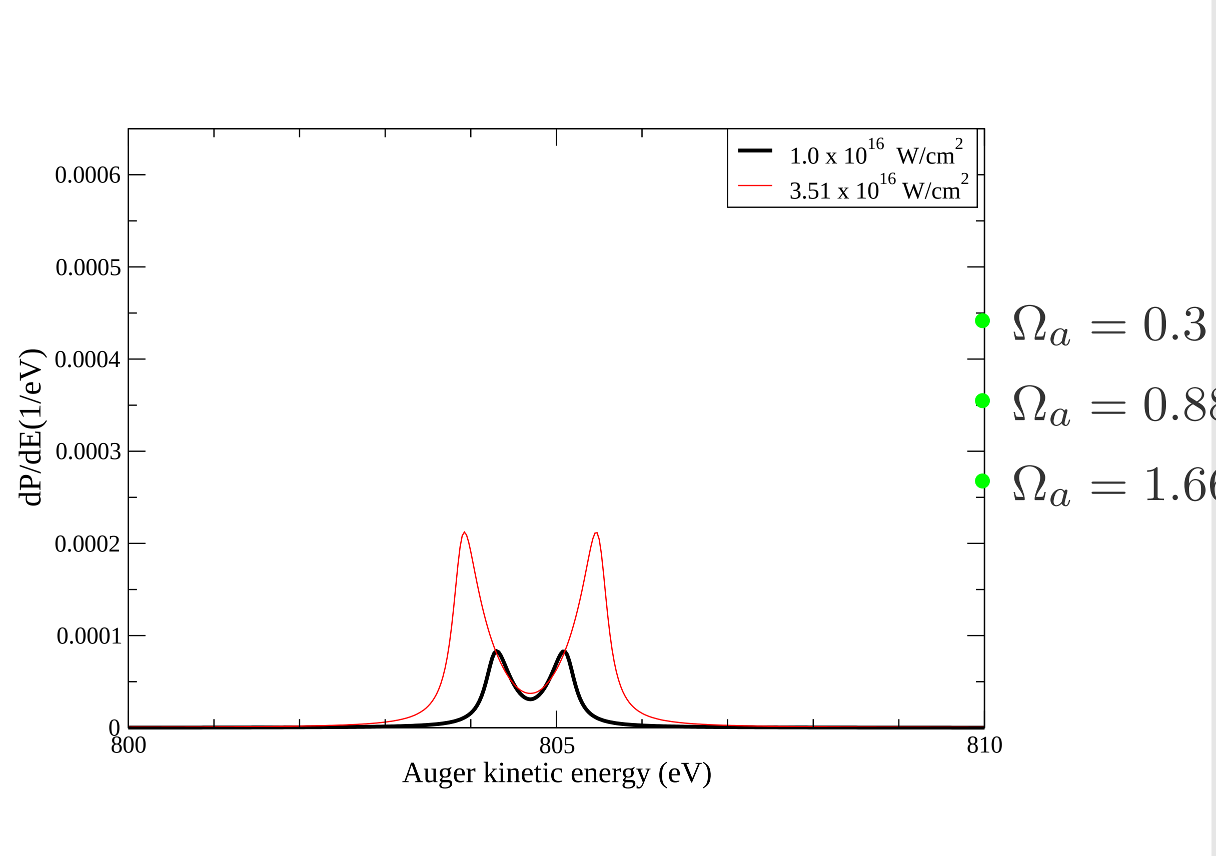

- $\Omega_a = 0.3 \sim \Gamma_i$
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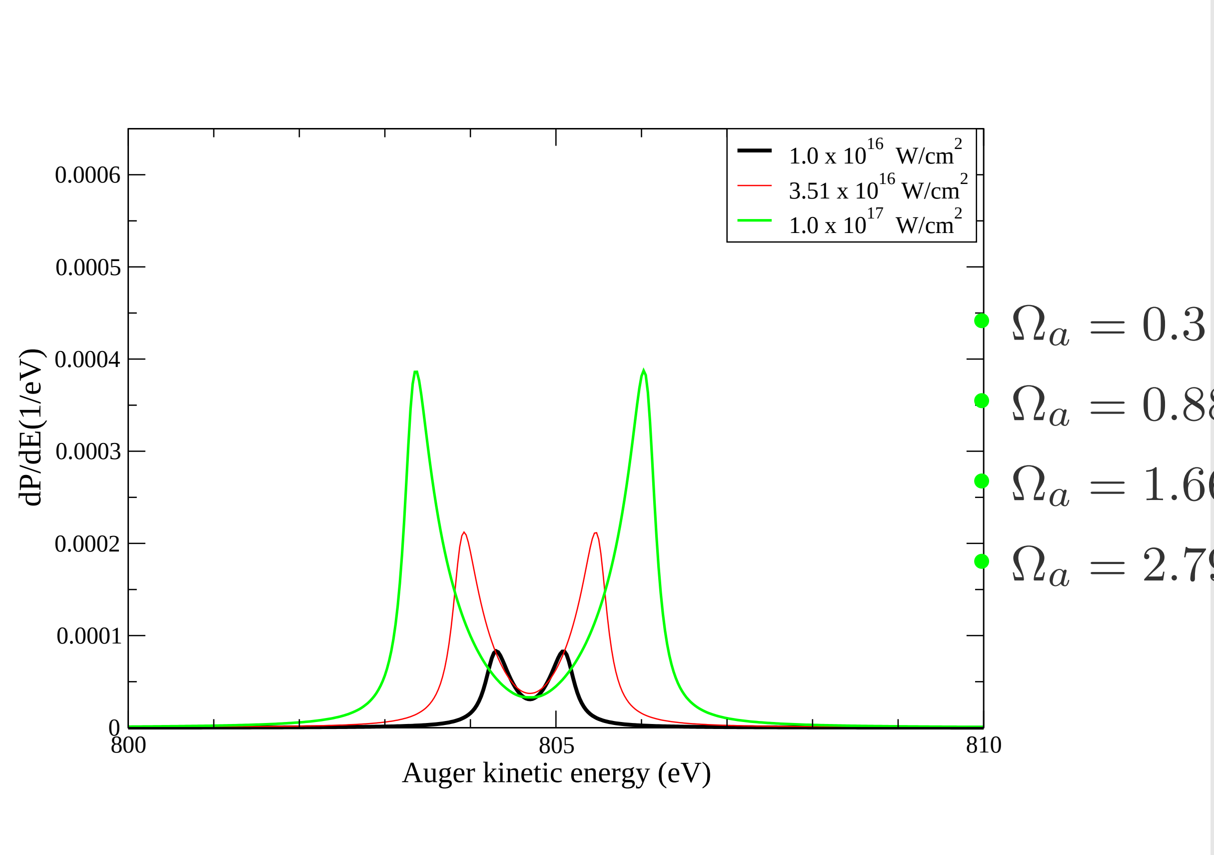
- $\Omega_a = 1.66 \sim 6\Gamma_i$ eV

Core resonant Auger kinetic spectra



- $\Omega_a = 0.3 \sim \Gamma_i$
- $\Omega_a = 0.88 \sim 3\Gamma_i$ eV
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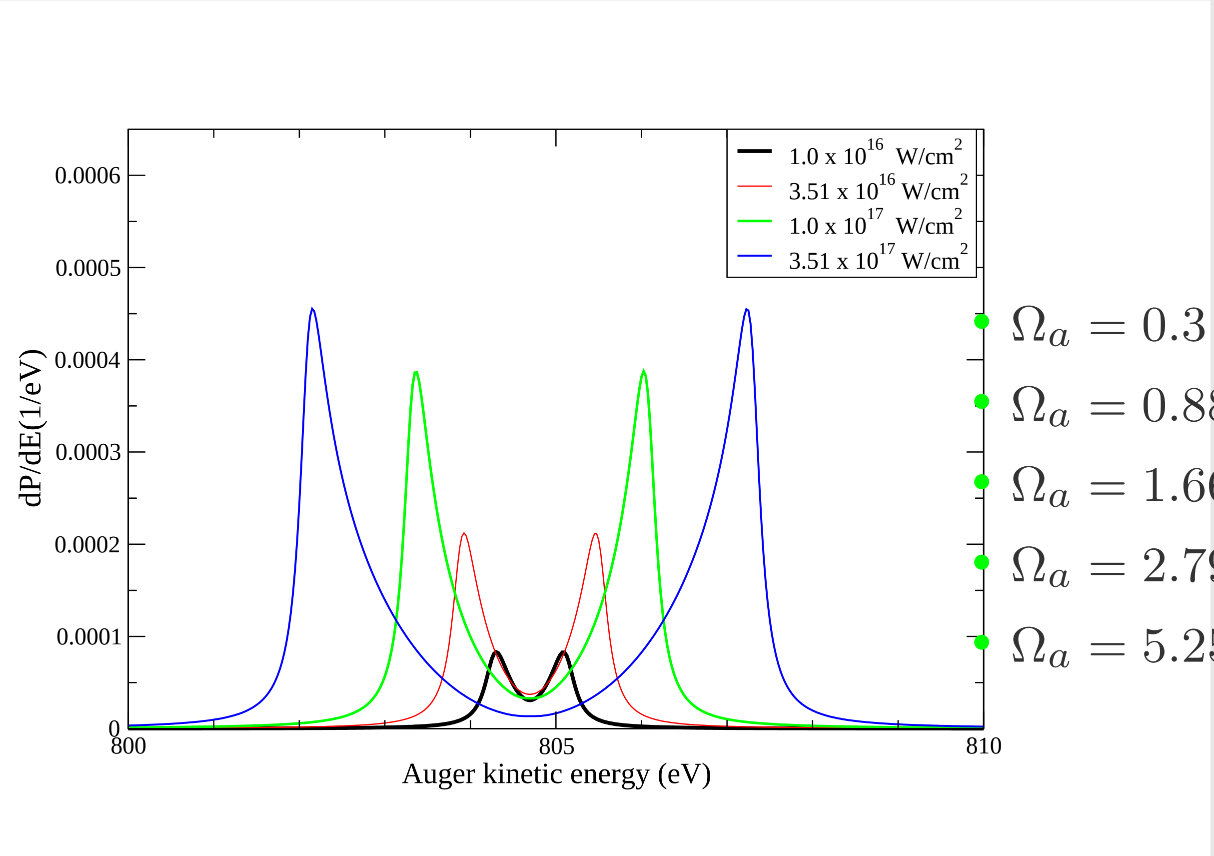

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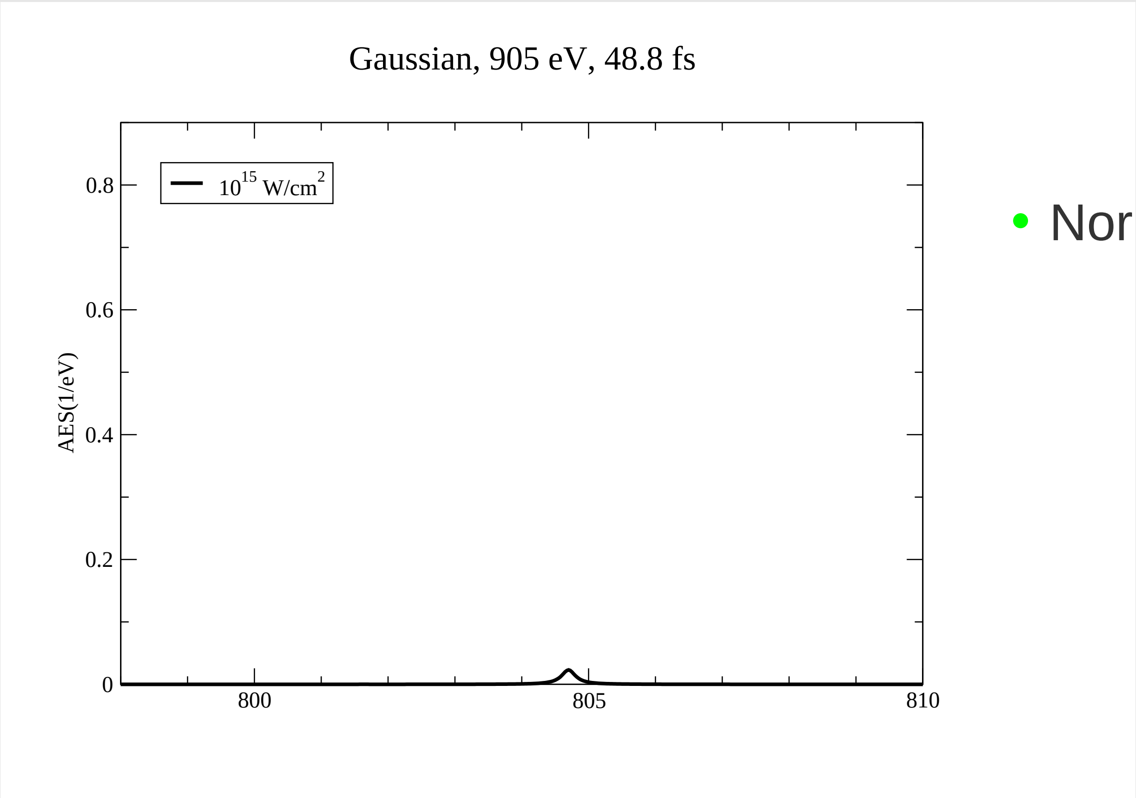
- $\Omega_a = 5.25 \sim 20\Gamma_i$ eV

DCU

Core-resonant Auger kinetic spectra 905 eV ($\delta_{a'} = 3$ eV)



- Normal Auger line

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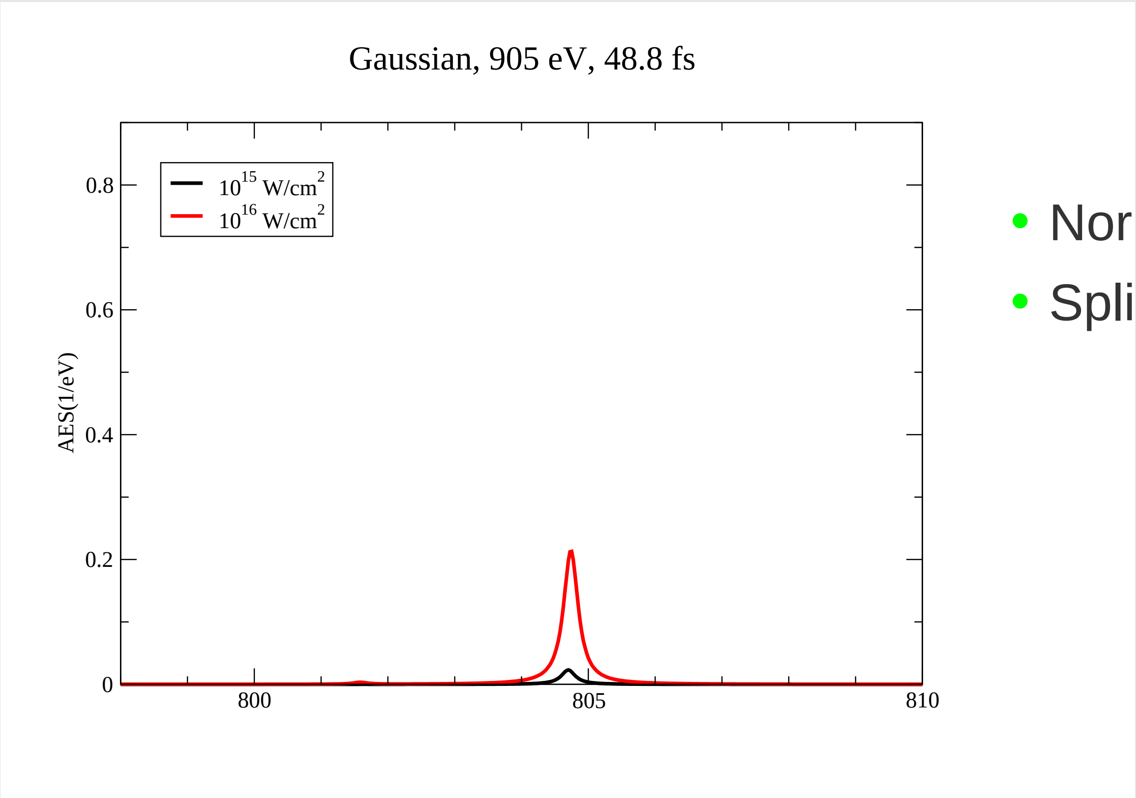

- Normal Auger line
- Splitting starts to appear

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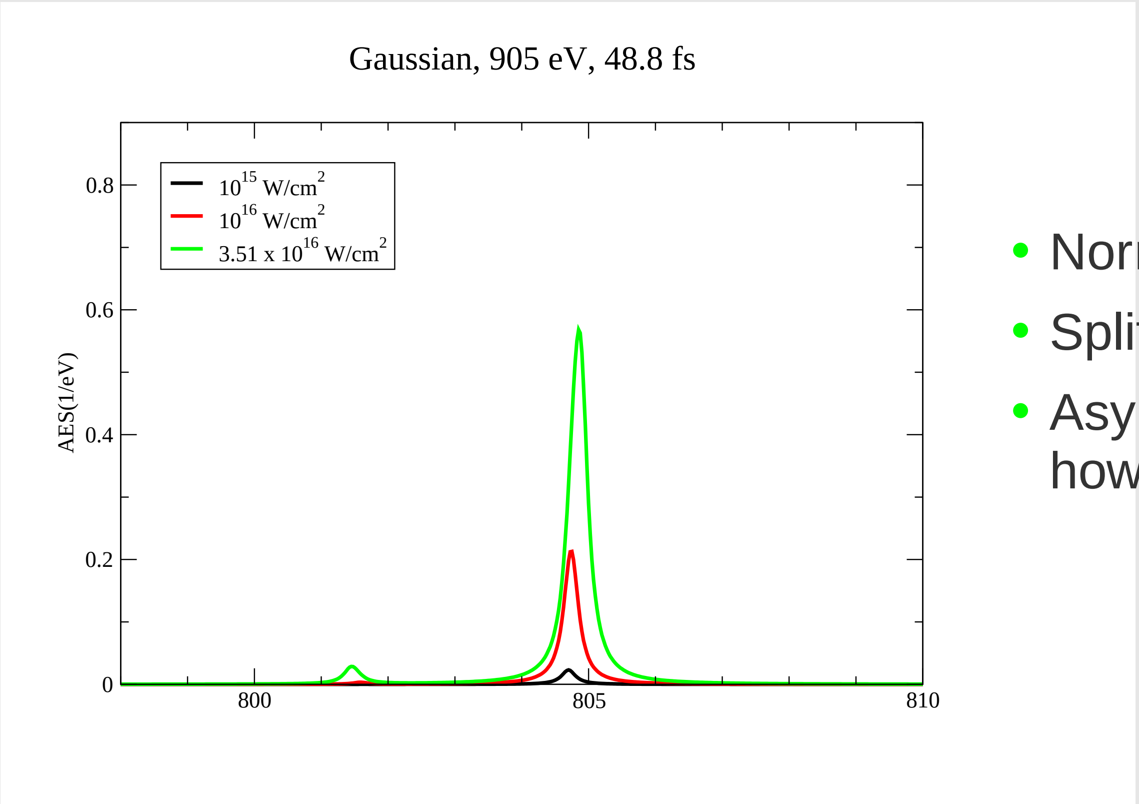

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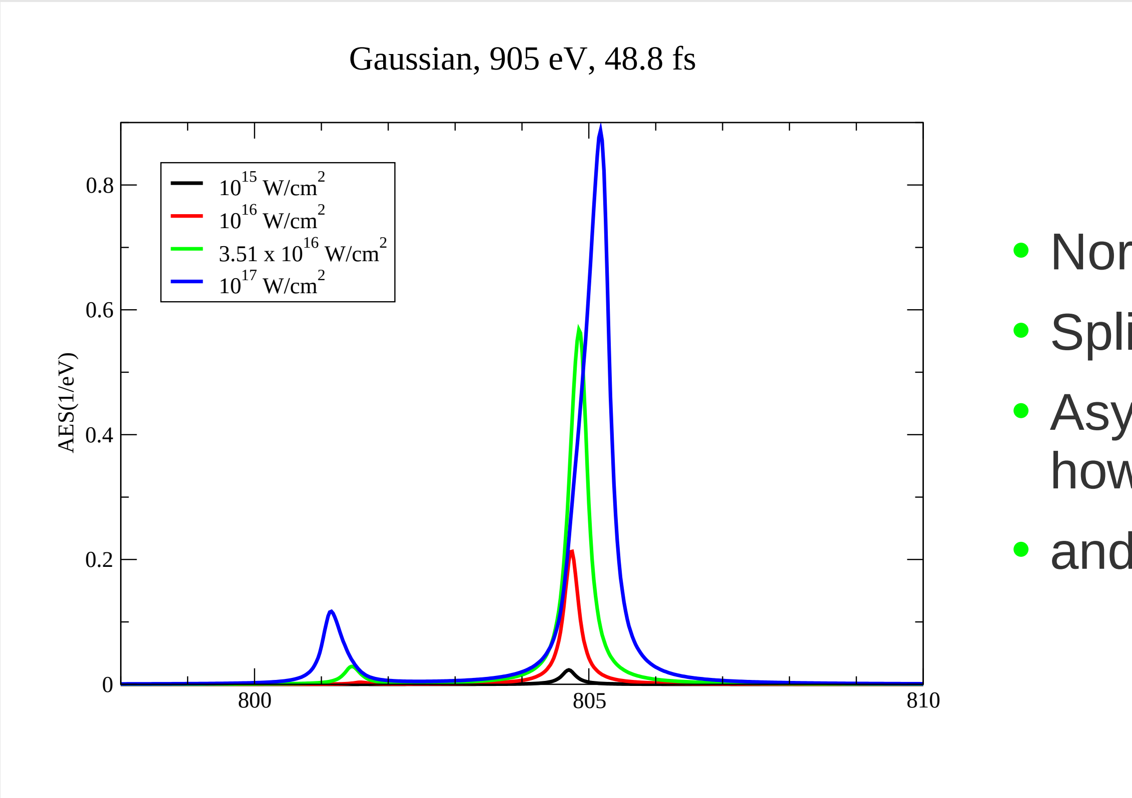

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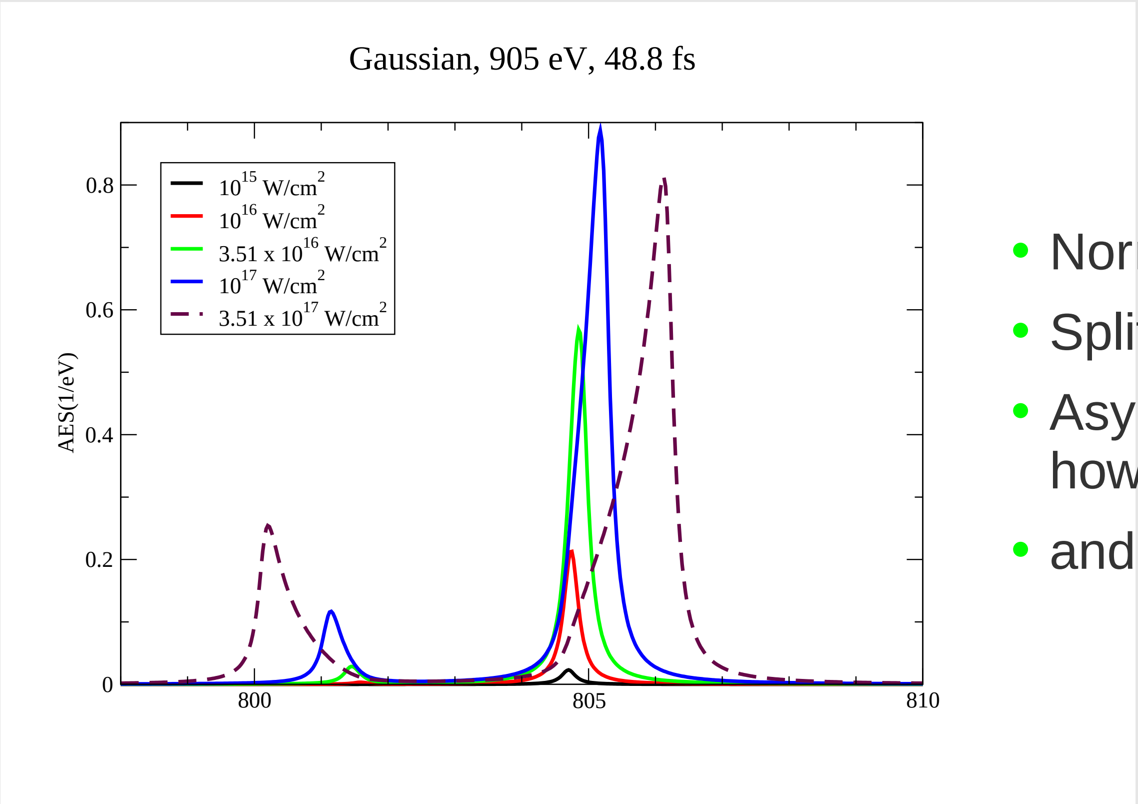

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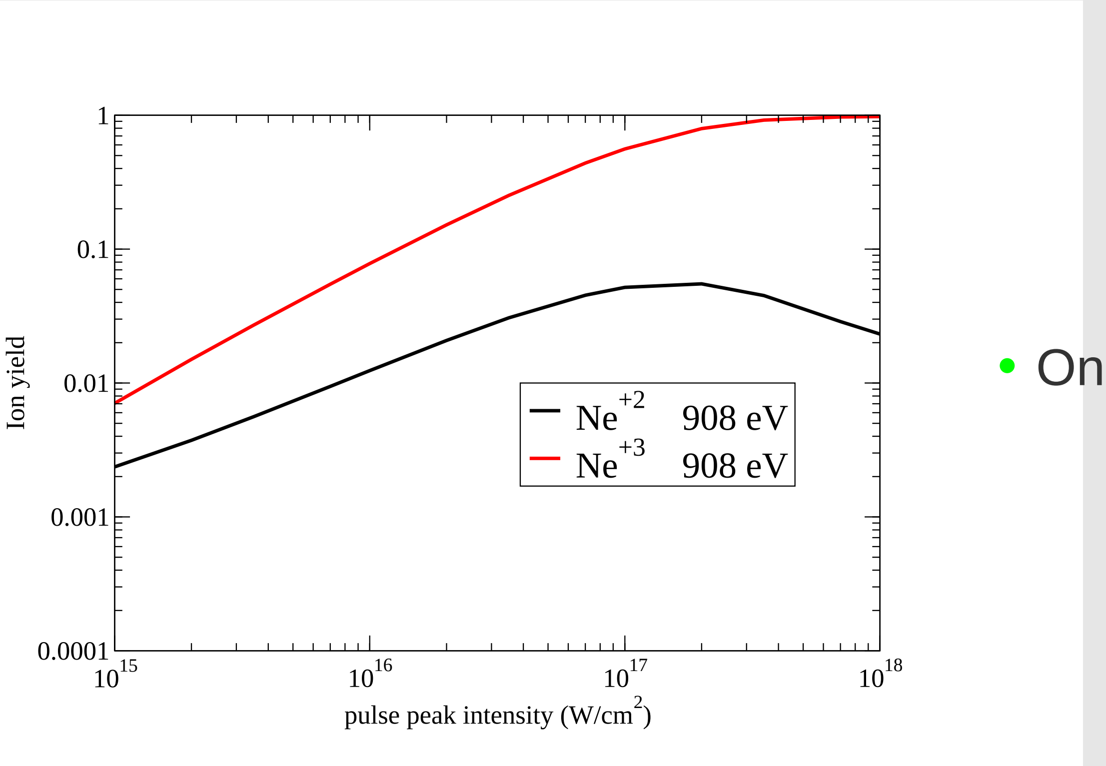
Ionization yields



- On resonance,

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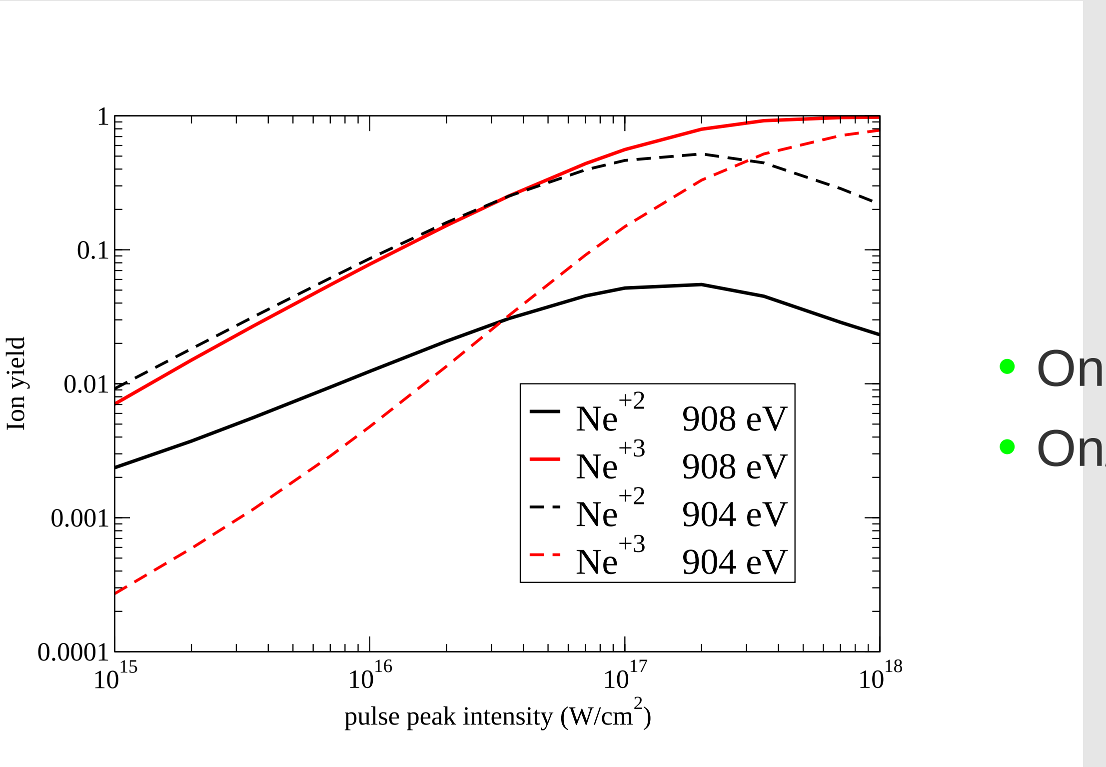

- On resonance,
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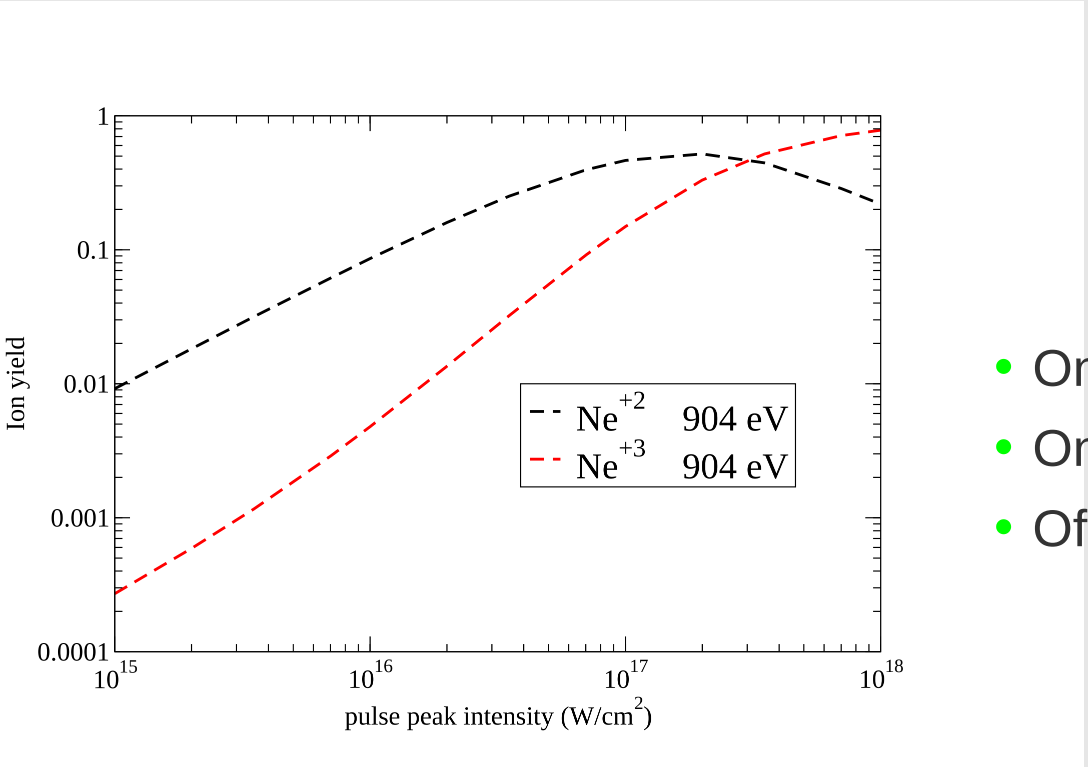

- On resonance,
- On/Off resonance
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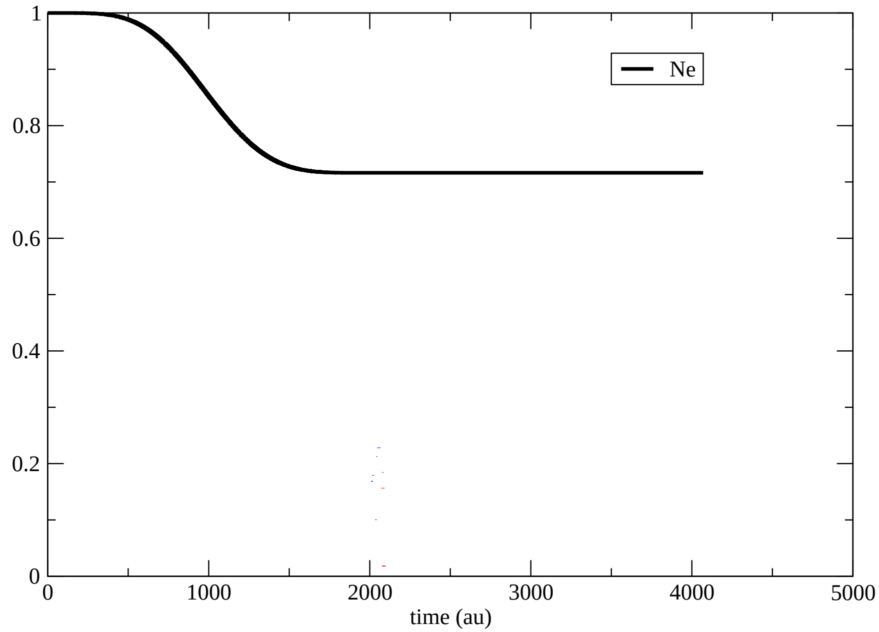
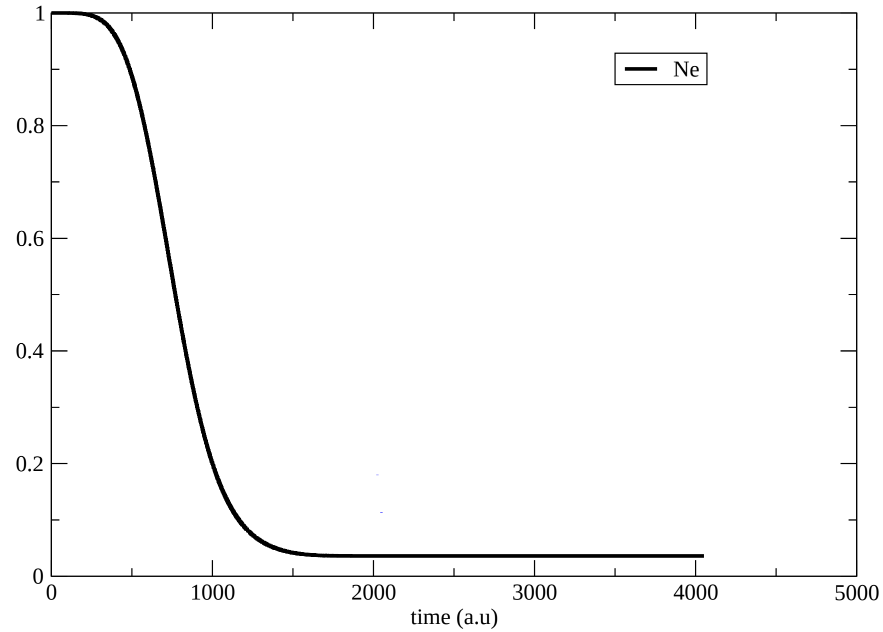
Time dependence of Ne, Ne^{+}, Ne^{+2}, $Ne^{+2}(1s^{-1} - 3p)$



$I_0 = 3.51 \times 10^{16}$W/cm^2



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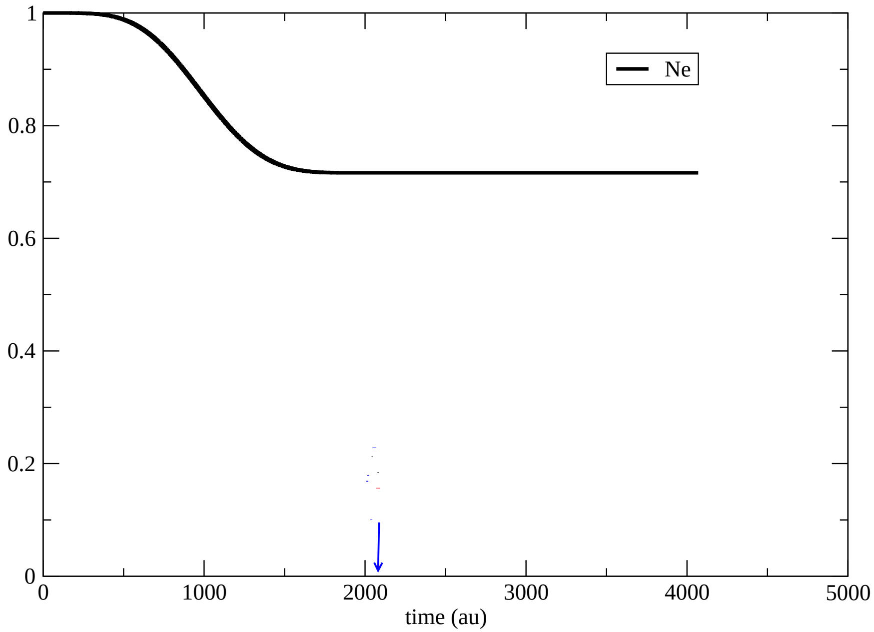

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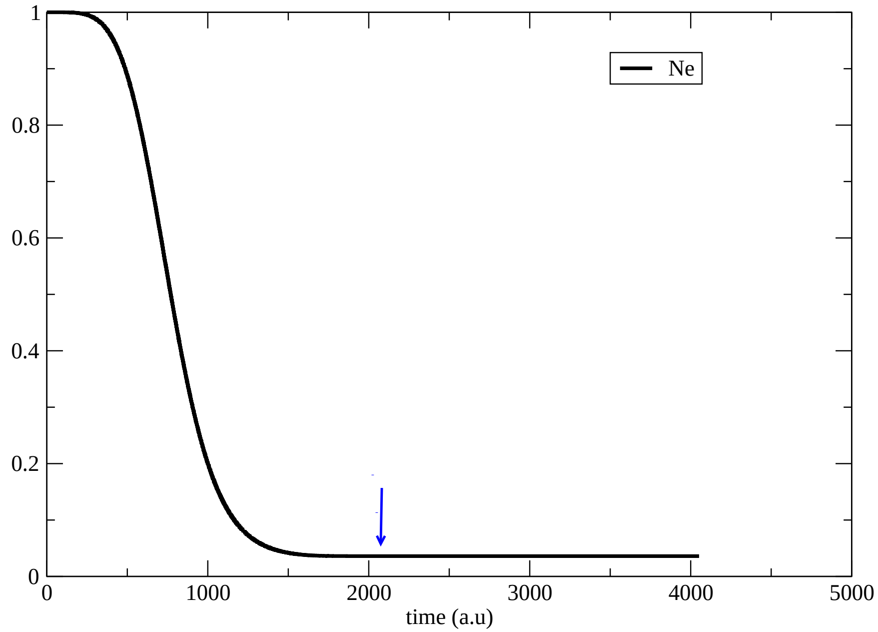

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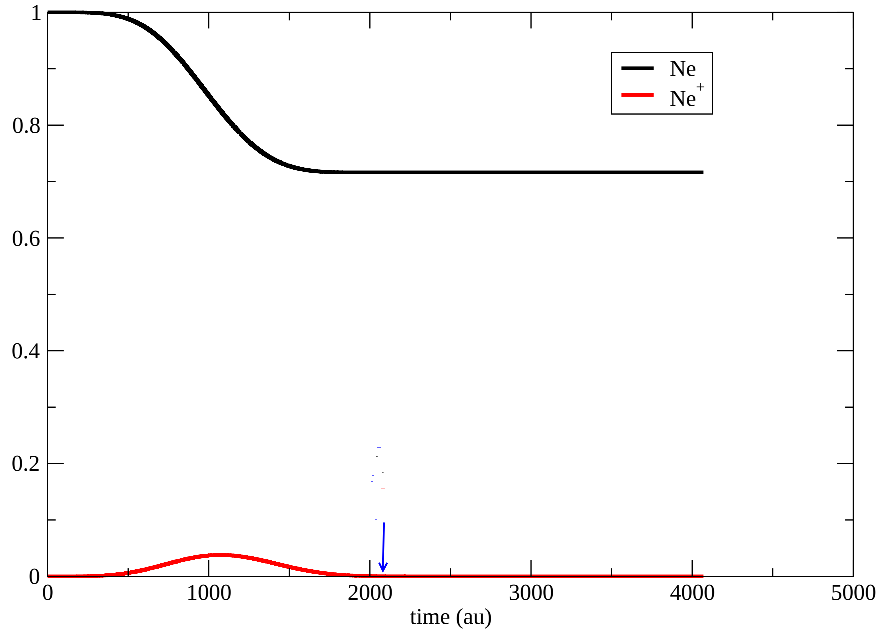

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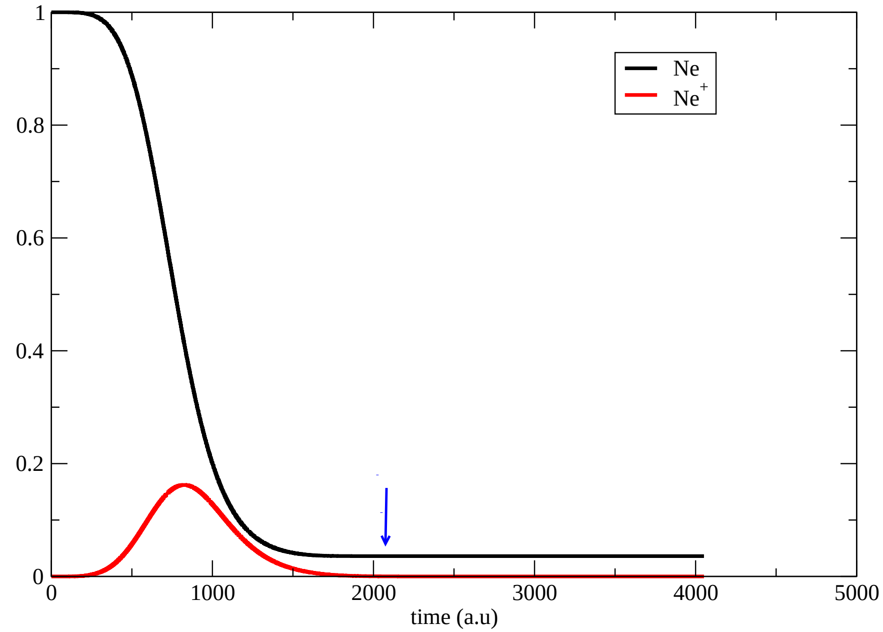

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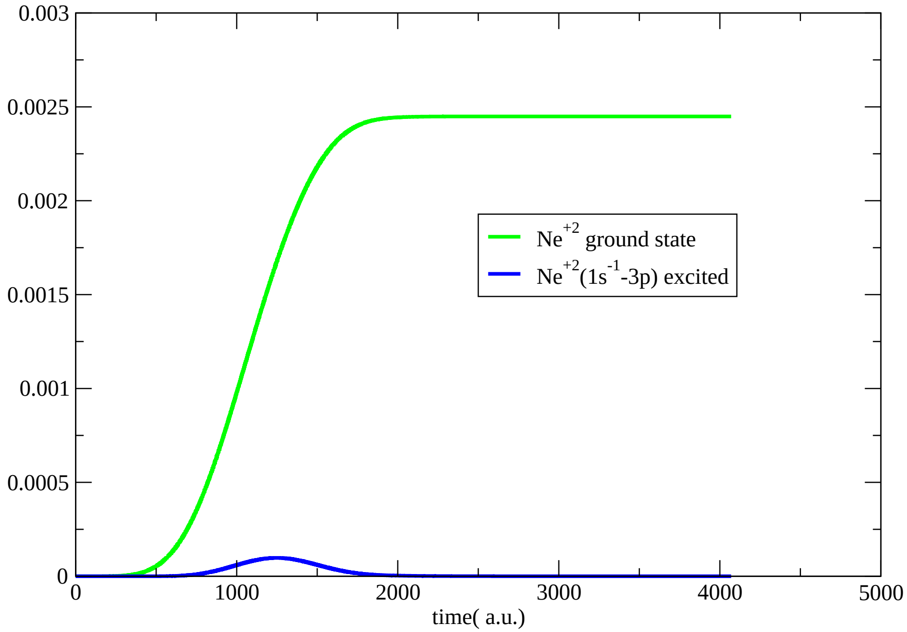

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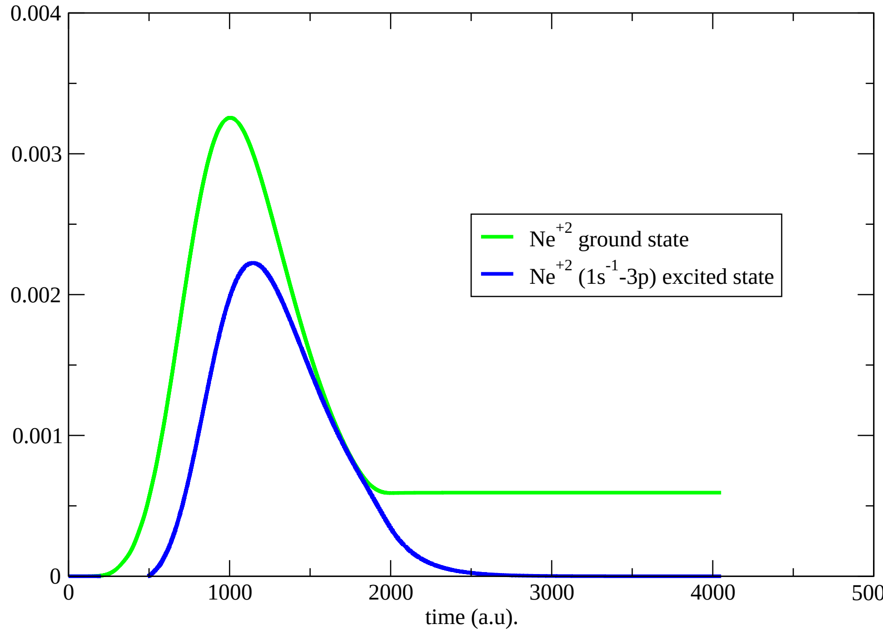

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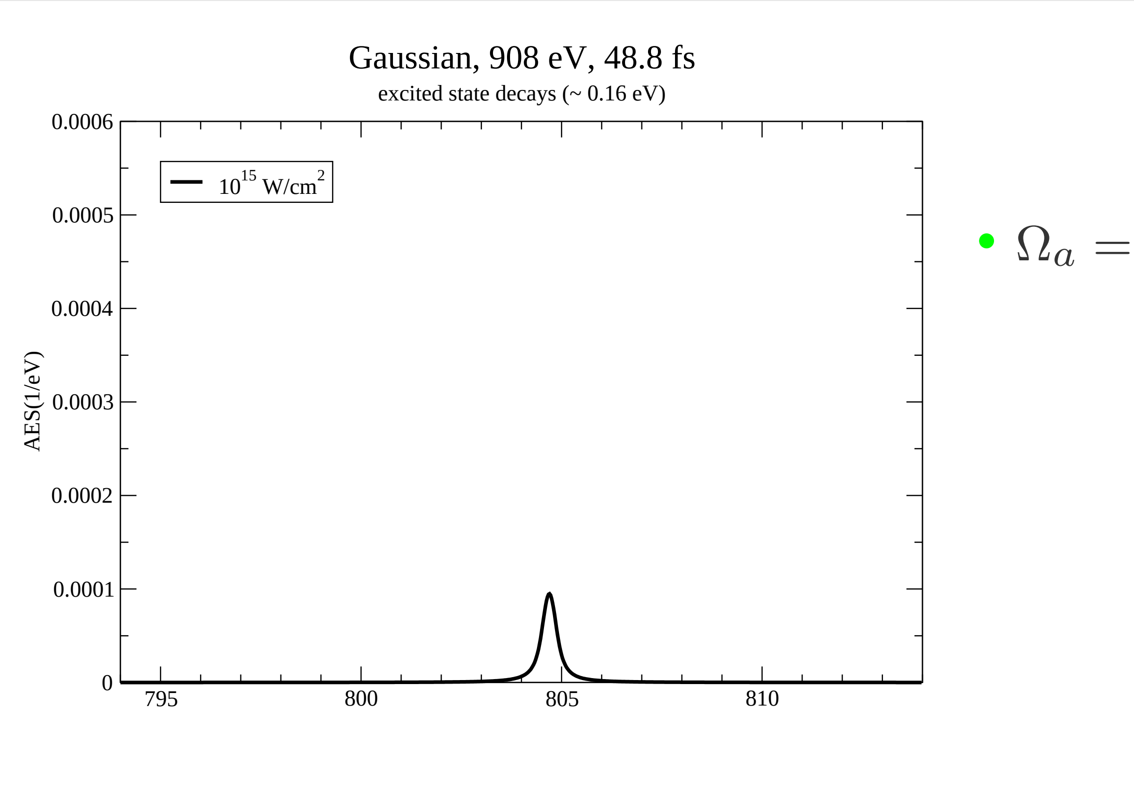
Core-resonant Auger kinetic spectra 908 eV ($\Gamma_{a'} = 0.16$ eV)



- $\Omega_a = 0.3$ eV

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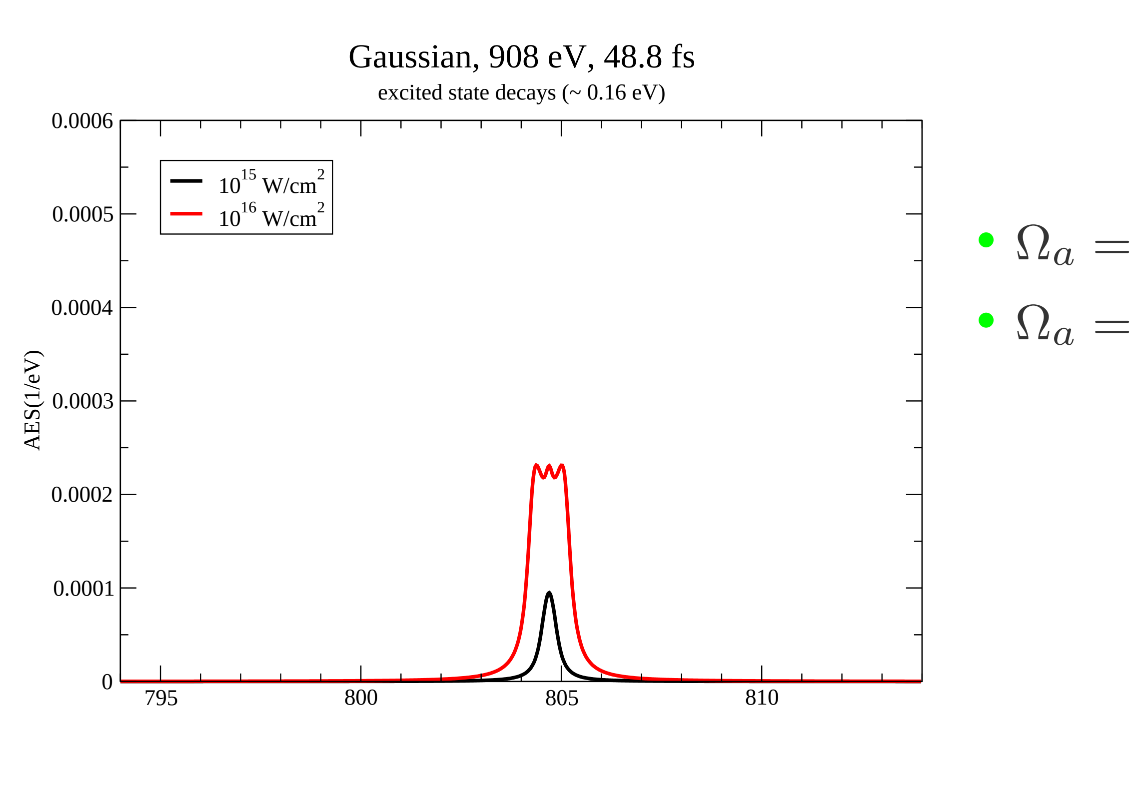

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DCU

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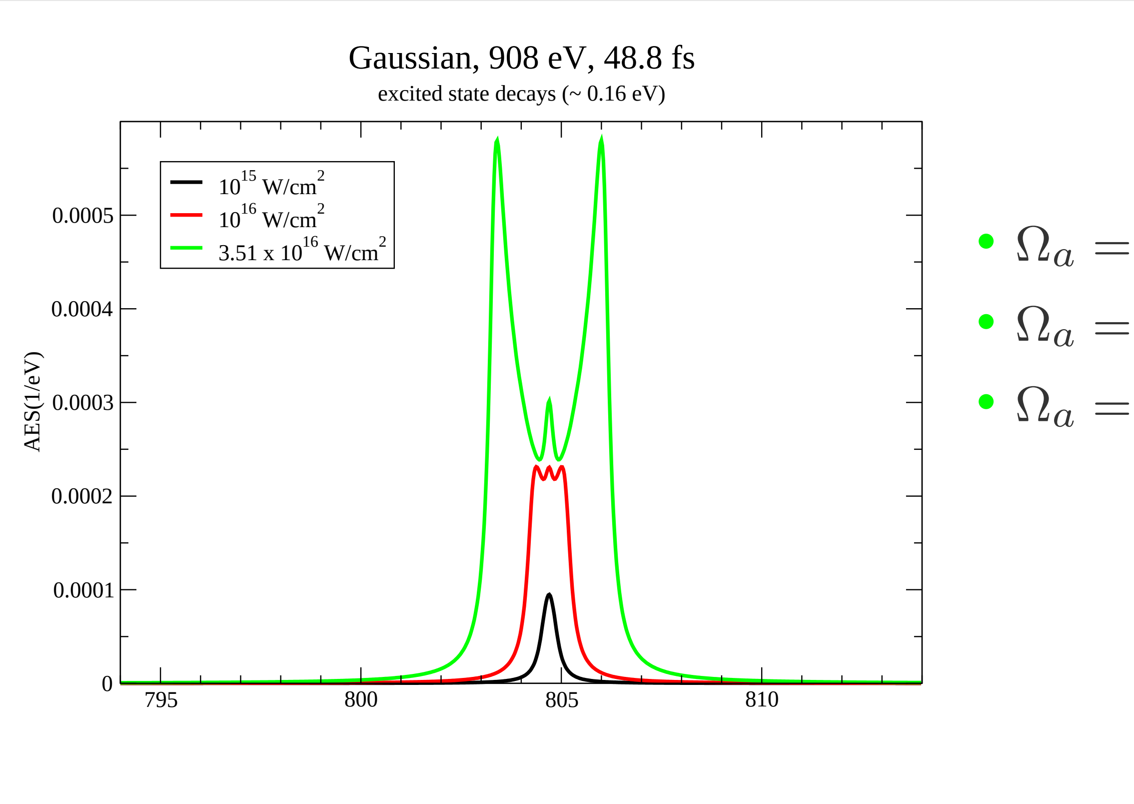

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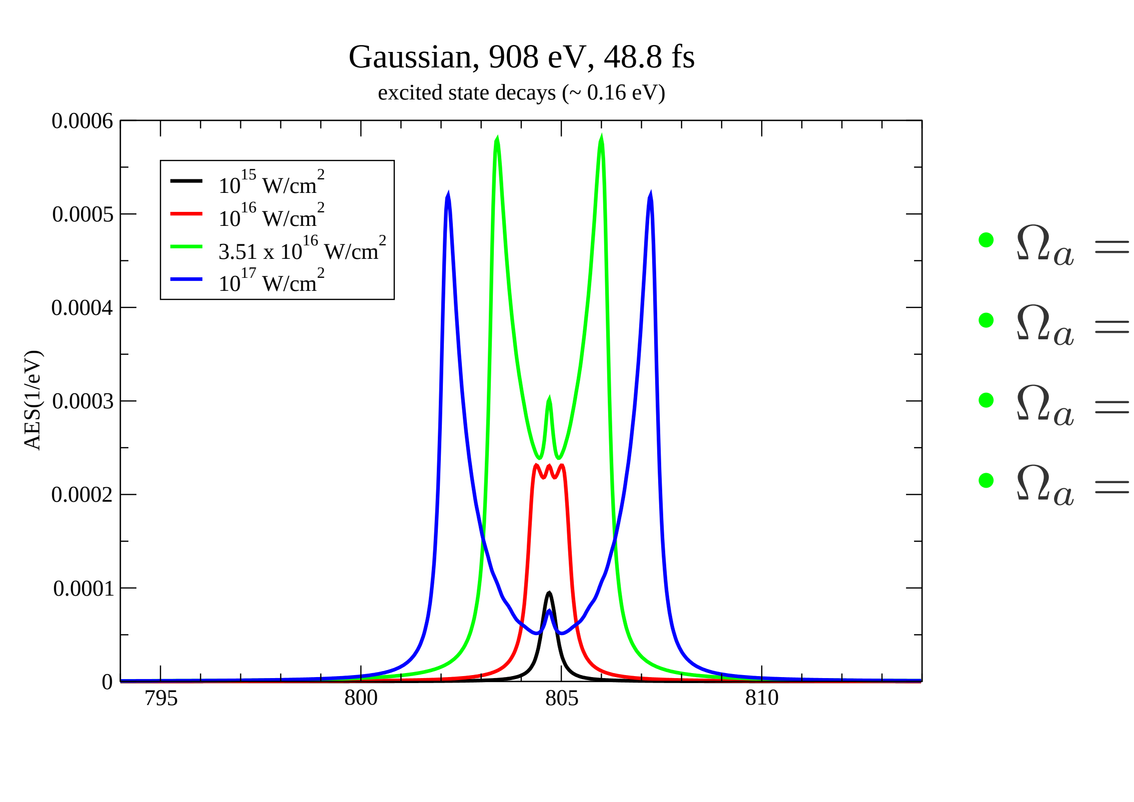

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DCU

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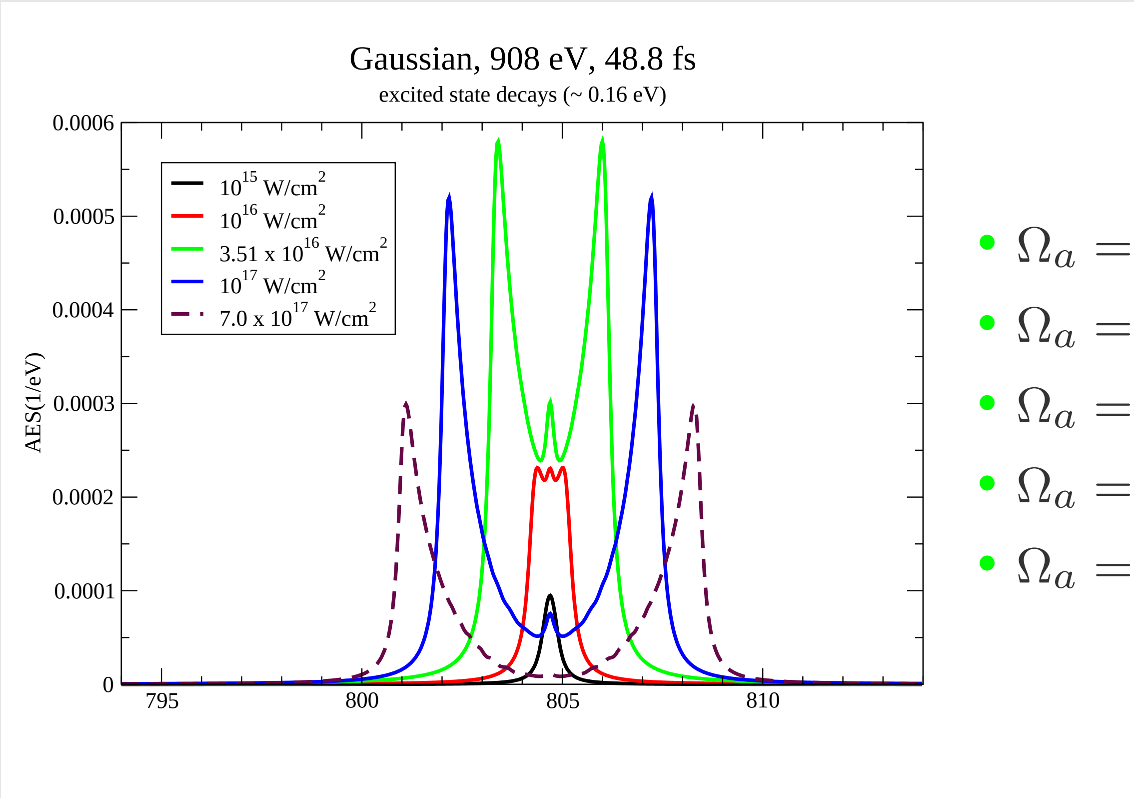

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DCU

Closer to experimental situation

DCU collaborators John Costello and Thomas (Mossy) Kelly are contributing on this part.

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- Field undergoes fluctuations
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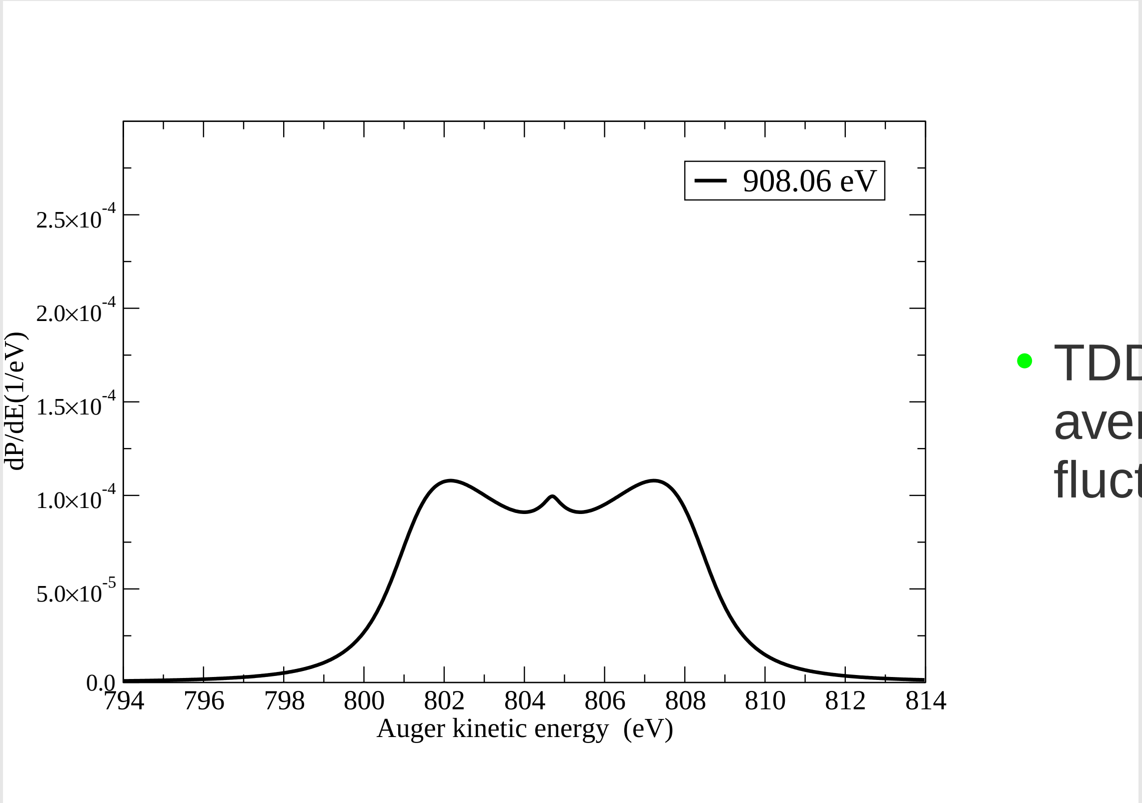
- All excited states of $Ne^{+2}(1s^{-1} - 3p)$ should be included
- The stochastic field fluctuations, will add it's bandwidth to the AES
- Volume integration of the AES is needed

Stochastic Pulse for 48.8 fs, xfel bandwidth = 4 eV



- TDDM Equations have averaged over the field fluctuations

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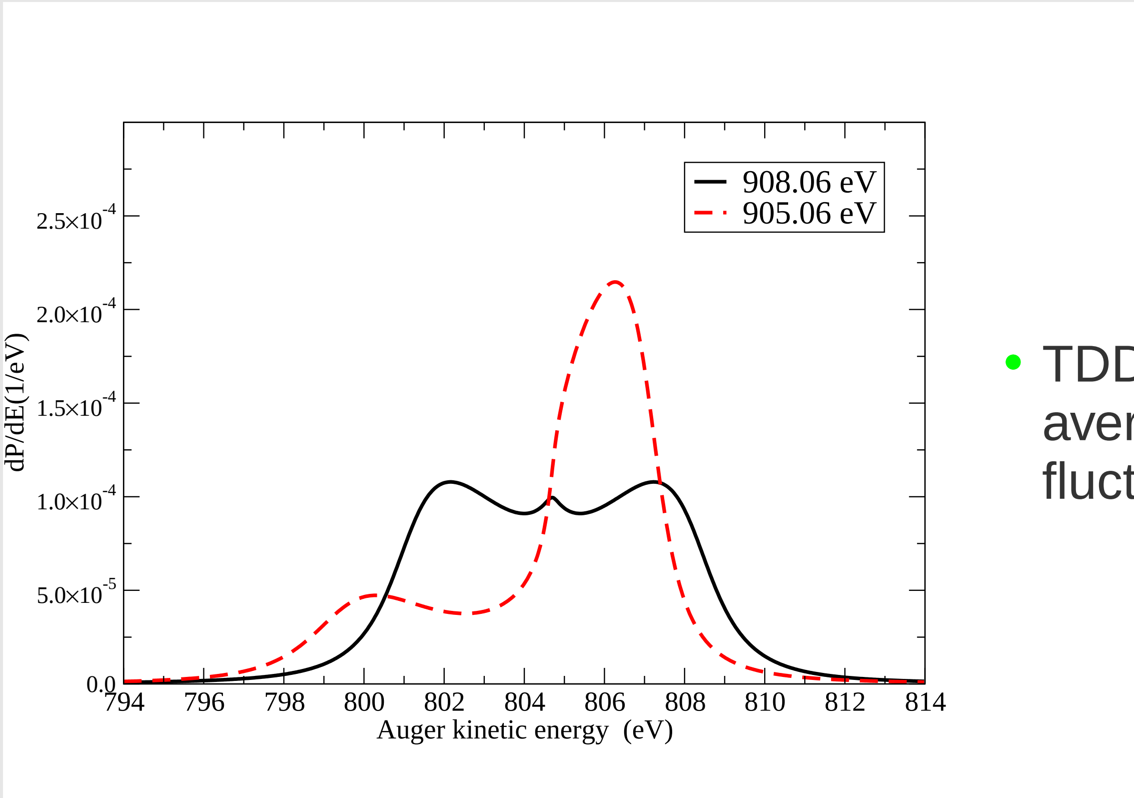

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Thanks for your attention

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