Positron annihilation on atomic core electrons + positive ions

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Motivation

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- Positron annihilation on core electrons is a key process in established experimental techniques:

  e.g. **P**ositron-induced **A**uger-**E**lectron **S**pectroscopy (PAES) [1]

  ⇓

  Real need for accurate core annihilation gamma-ray-spectra and probabilities - Correlations??!

- Discrepancies between theory [2] and measured [3] γ-spectra of noble gases: due to core annihilation?

- Positive ions: limited theory, no experiments!

[1] Weiss et al., Phys. Rev. Lett. **61**, 2245 (1988).
[2] Dunlop and Gribakin, J. Phys. B. **39** 1647 (2005).
[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997).

Fundamental questions

1) What effect do correlations have on γ-spectra?

- Noble gas atoms: Ar, Kr, Xe
- H-like positive ions: He^{+}, Li^{2+}, B^{4+}, F^{8+} (ionization energies span those typical of core electrons)

2) Many-electron atoms:
importance of core contribution to γ-spectra?

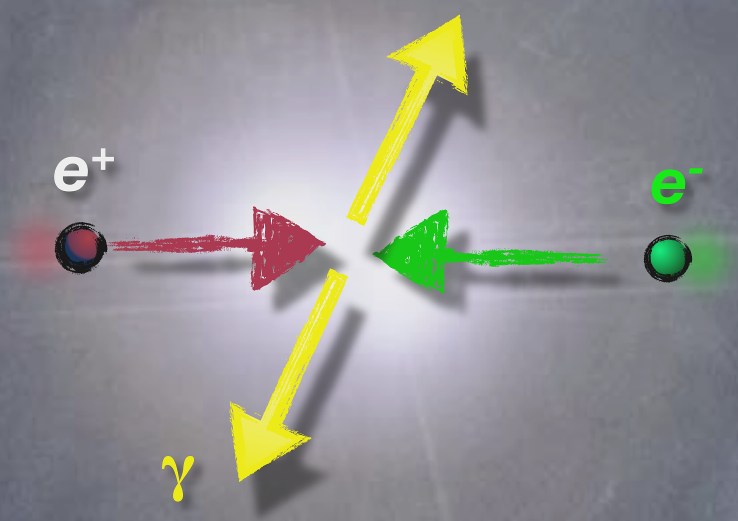
Outline

1. Many-body theory of annihilation γ-spectra
2. Results: γ-spectra of nobles + positive ions
   - i) Effect of correlations
   - ii) Core contribution + comparison with experiment

Positron annihilation gamma-spectra

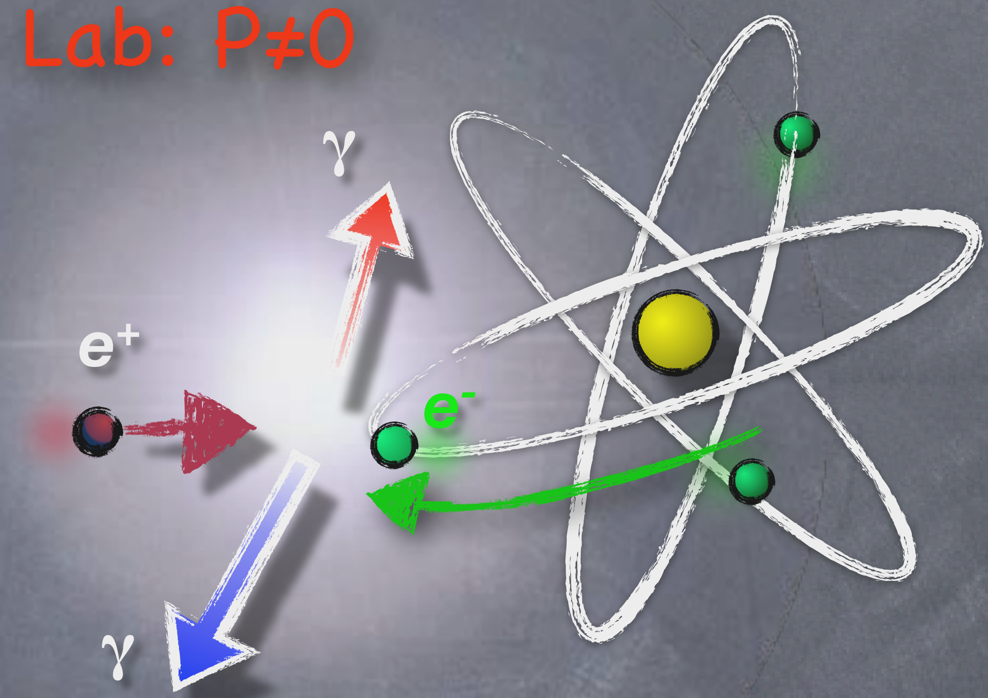
Low-energy positrons e.g., thermal: k≈0.04au

COM frame: **P**=0



$E_\gamma \simeq mc^2 = 511$ keV

Lab: P≠0



$E_\gamma = mc^2 + \epsilon$

Doppler shift: $\epsilon \sim$ several keV.

γ-spectra: $w_n(\epsilon)$ shape characteristic of electron state n

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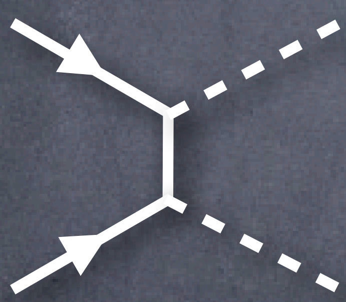
Many-body theory of γ-spectra

γ-spectrum : $$w_{n\varepsilon}(\epsilon) = \frac{1}{c}\int_{2|\epsilon|/c}^{\infty} |A_{n\varepsilon}(\mathbf{P})|^2 \frac{d\Omega_{\mathbf{P}}}{4\pi} P dP$$

Ann. amplitude:

$$A_{n\varepsilon}(\mathbf{P}) = \langle \Psi_n^{N-1} | \hat{O}_a(\mathbf{P}) | \Psi_\varepsilon^{N+1} \rangle$$



$$\hat{O}_a(\mathbf{P}) \propto \int d^3r \, e^{-i\mathbf{P}\cdot\mathbf{r}} \hat{\psi}(\mathbf{r})\hat{\varphi}(\mathbf{r})$$

NR annihilation: occurs at single-point!

Dyson expansion in: $H_{\rm int} = \hat{V}_{\rm e^+e^-} + \hat{V}_{\rm e^-e^-} - \hat{U}_{\rm HF}$

using Hatree-Fock as starting approximation...

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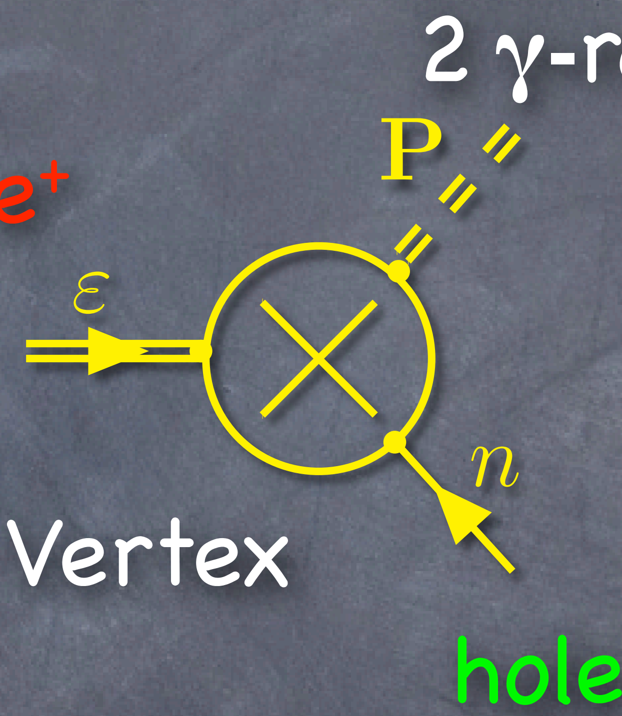
Many-body theory: annihilation amplitude

γ-spectrum : $$w_{n\varepsilon}(\epsilon) = \frac{1}{c}\int_{2|\epsilon|/c}^{\infty} |A_{n\varepsilon}(\mathbf{P})|^2 \, \frac{d\Omega_{\mathbf{P}}}{4\pi} P dP$$

General diagram:

$$A_{n\varepsilon}(\mathbf{P}) =$$



Different approximations to:

1) Vertex

2) Positron wavefunction

Many-body theory: annihilation amplitude

γ-spectrum :
$$w_{n\varepsilon}(\epsilon) = \frac{1}{c}\int_{2|\epsilon|/c}^{\infty} |A_{n\varepsilon}(\mathbf{P})|^2 \, \frac{d\Omega_{\mathbf{P}}}{4\pi} P dP$$

General diagram:

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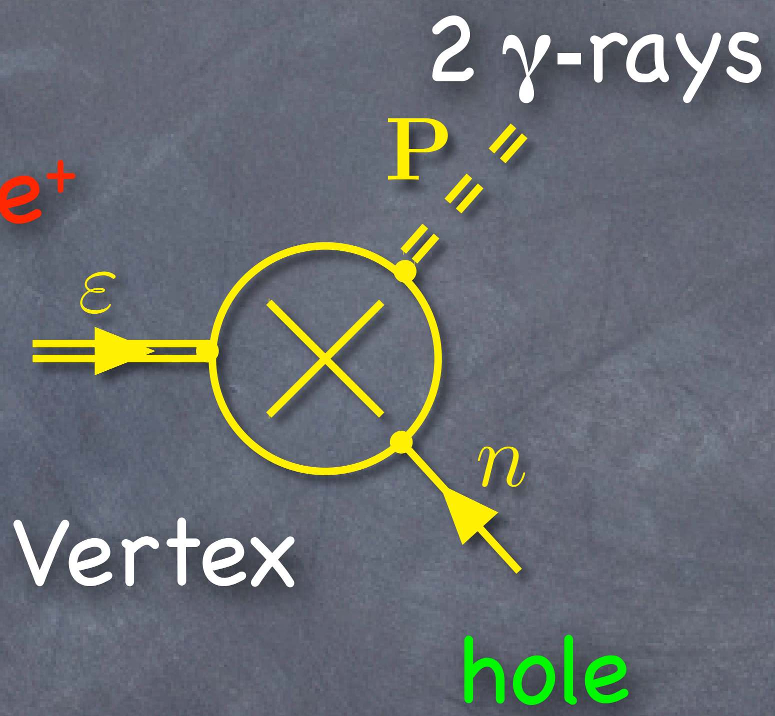
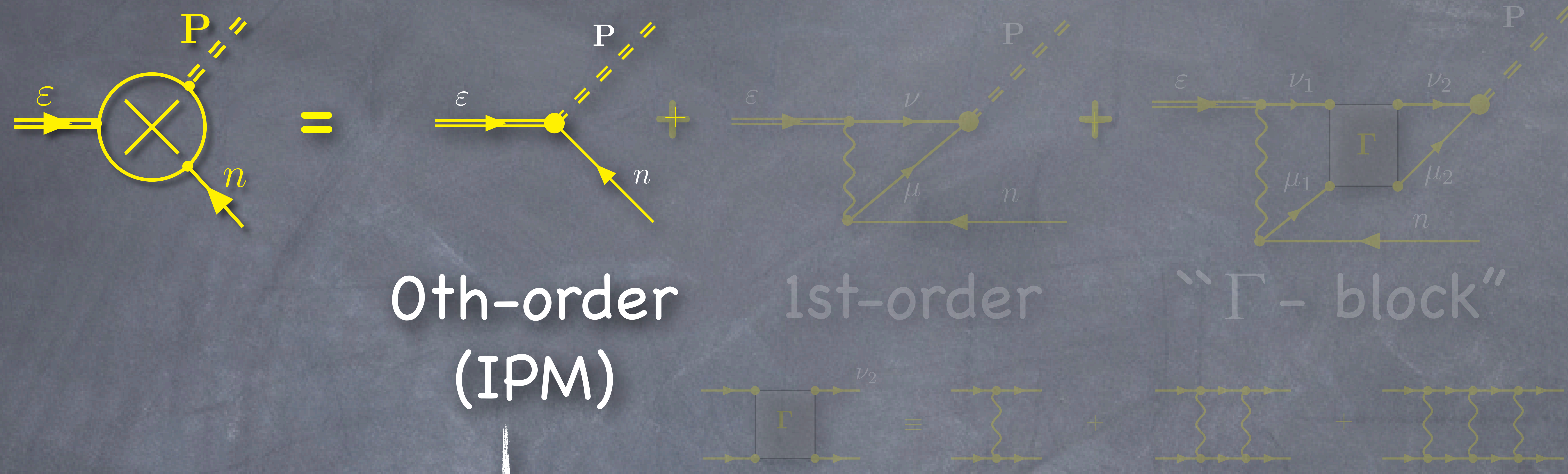


Different approximations to:

1) Vertex

2) Positron wavefunction

Annihilation amplitude: VERTEX



e+ hole

$$A_{n\varepsilon}(\mathbf{P}) = \int e^{-i\mathbf{P}\cdot\mathbf{r}} \psi_\varepsilon(\mathbf{r})\varphi_n(\mathbf{r})\, d\mathbf{r}$$

$$+ \int e^{-i\mathbf{P}\cdot\mathbf{r}} \psi_\varepsilon(\mathbf{r}_1)\tilde{\Delta}_{\mathbf{P}}(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r})\varphi_n(\mathbf{r}_2)\, d\mathbf{r}_1 d\mathbf{r}_2\, d\mathbf{r}$$

non-local vertex corrections:
describe short-range electron-positron interactions

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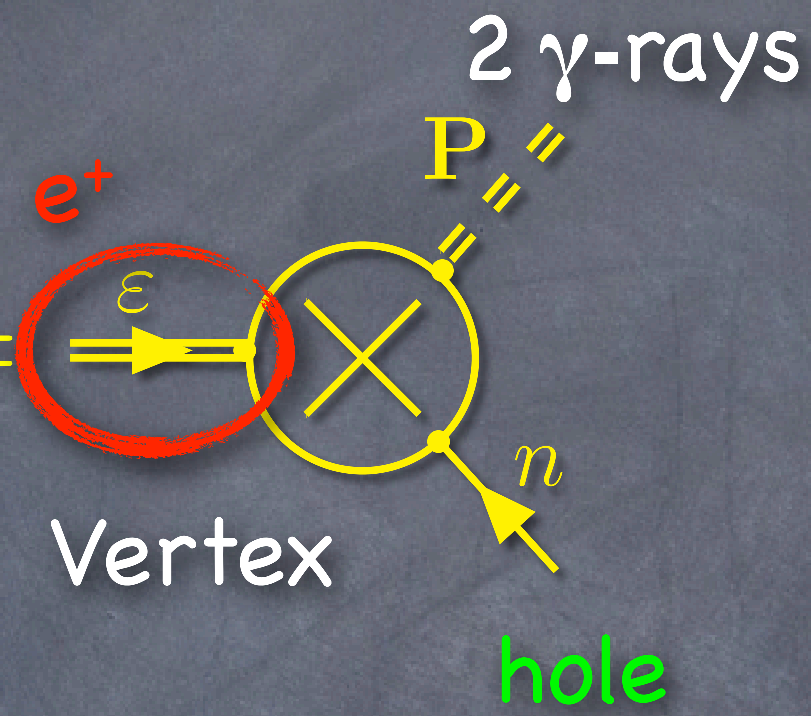
Annihilation amplitude: POSITRON WAVEFUNCTION

General diagram:

$$A_{n\varepsilon}(\mathbf{P}) =$$



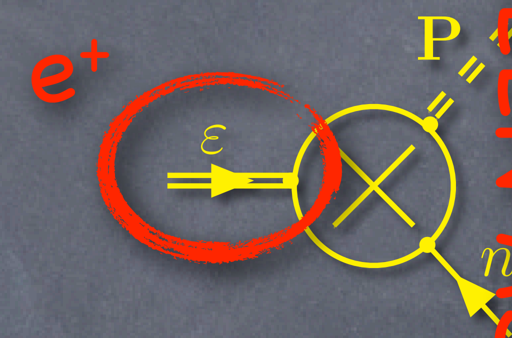
Different approximations to:

1) Vertex

2) Positron wavefunction

Annihilation amplitude: POSITRON WAVEFUNCTION

Two approximations: (1) Hatree-Fock; (2) "Dyson"

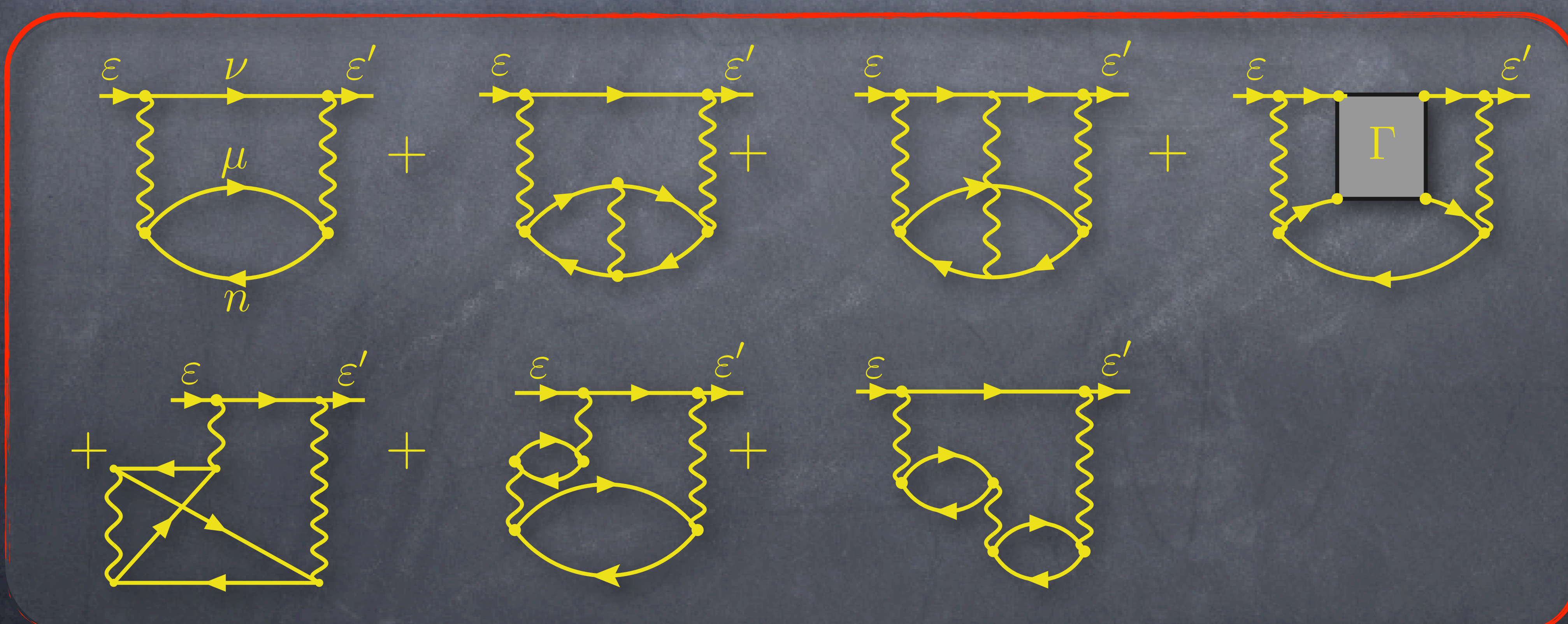


Dyson-equation:

$$(H_0 + \Sigma_\varepsilon)\psi_\epsilon(\mathbf{r}) = \varepsilon\psi_\epsilon(\mathbf{r})$$

Hartree-Fock

$$\hat{\Sigma}_\varepsilon\psi_\varepsilon(\mathbf{r}) \equiv \int \Sigma_\varepsilon(\mathbf{r},\mathbf{r}')\psi_\varepsilon(\mathbf{r}')d\mathbf{r}'$$



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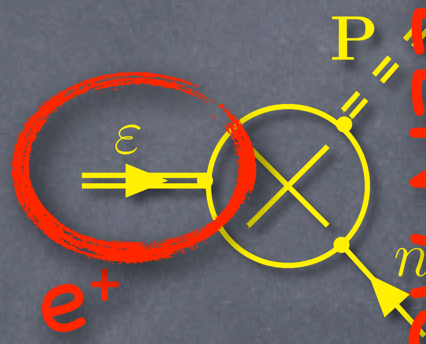
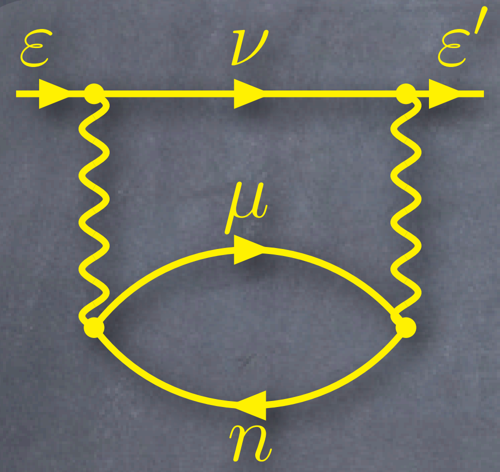
Annihilation amplitude: POSITRON WAVEFUNCTION



$$(H_0 + \Sigma_\varepsilon)\psi_\epsilon(\mathbf{r}) = \varepsilon\psi_\epsilon(\mathbf{r})$$

e.g., polarization of atom:



$$= \sum_{\mu,\nu} \frac{\langle \varepsilon' n | V | \mu, \nu \rangle \langle \nu, \mu | V | n \varepsilon \rangle}{(\varepsilon + \varepsilon_n - \varepsilon_\mu - \varepsilon_\nu)}$$

...attractive for small positron energy.

$$\lim_{r\to\infty} \Sigma^{(2)} \simeq -\frac{\alpha_d}{2r^4}\delta(\mathbf{r}-\mathbf{r}'), \quad \text{local!}$$

where α_d is the static dipolarizability of the atom.

Outline

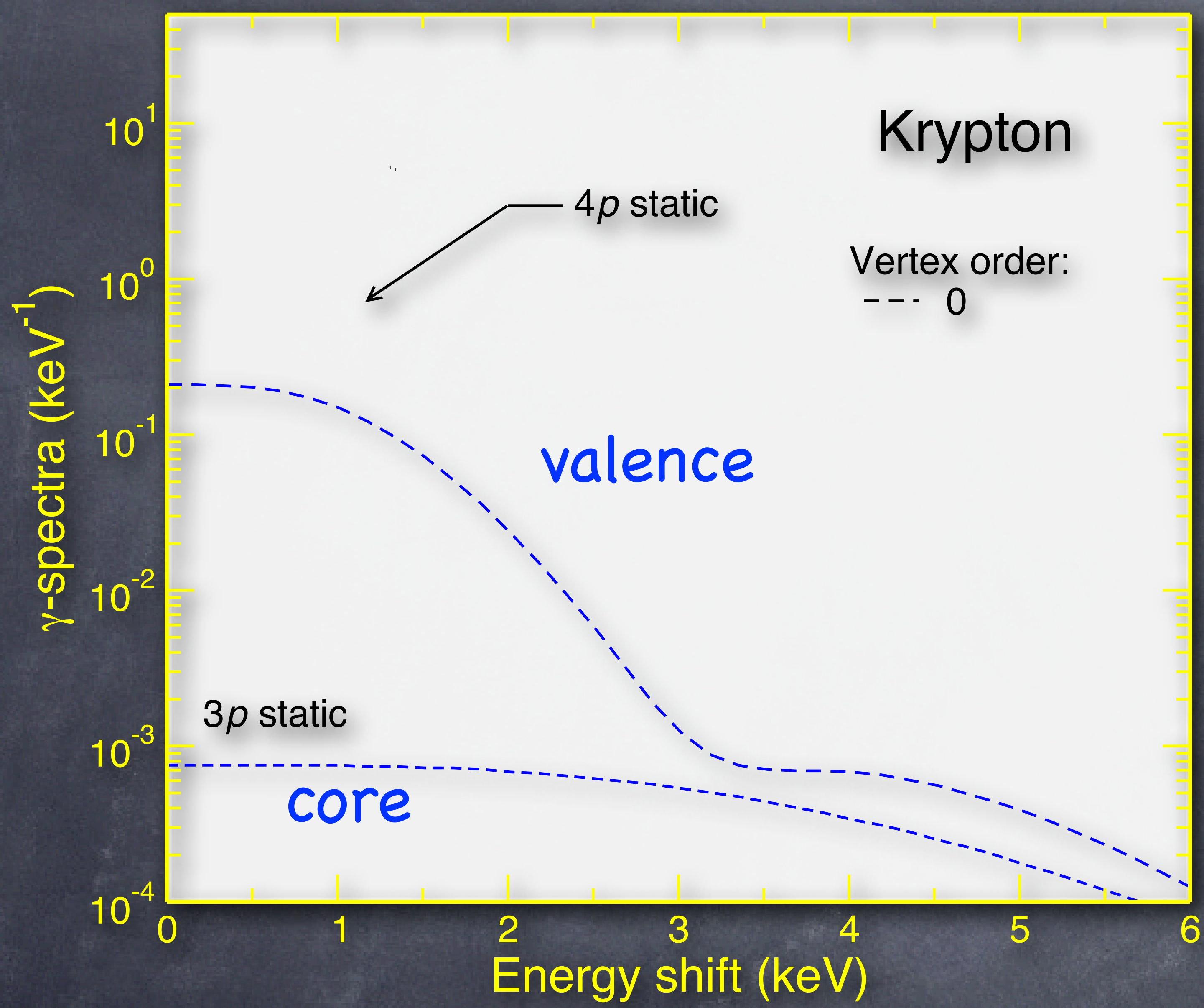
1. Many-body theory of annihilation γ-spectra

2. Results: γ-spectra of nobles + positive ions

   i) Effect of correlations

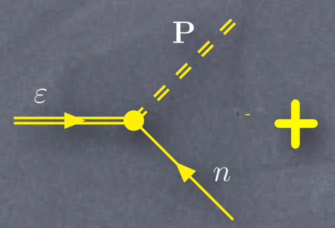
   ii) Core contribution + comparison with experiment

Effect of correlations: (1) VERTEX



$$\lambda_{\mathrm{ann.}} \propto \int w(\varepsilon) d\varepsilon$$

VERTEX ENHANCEMENT



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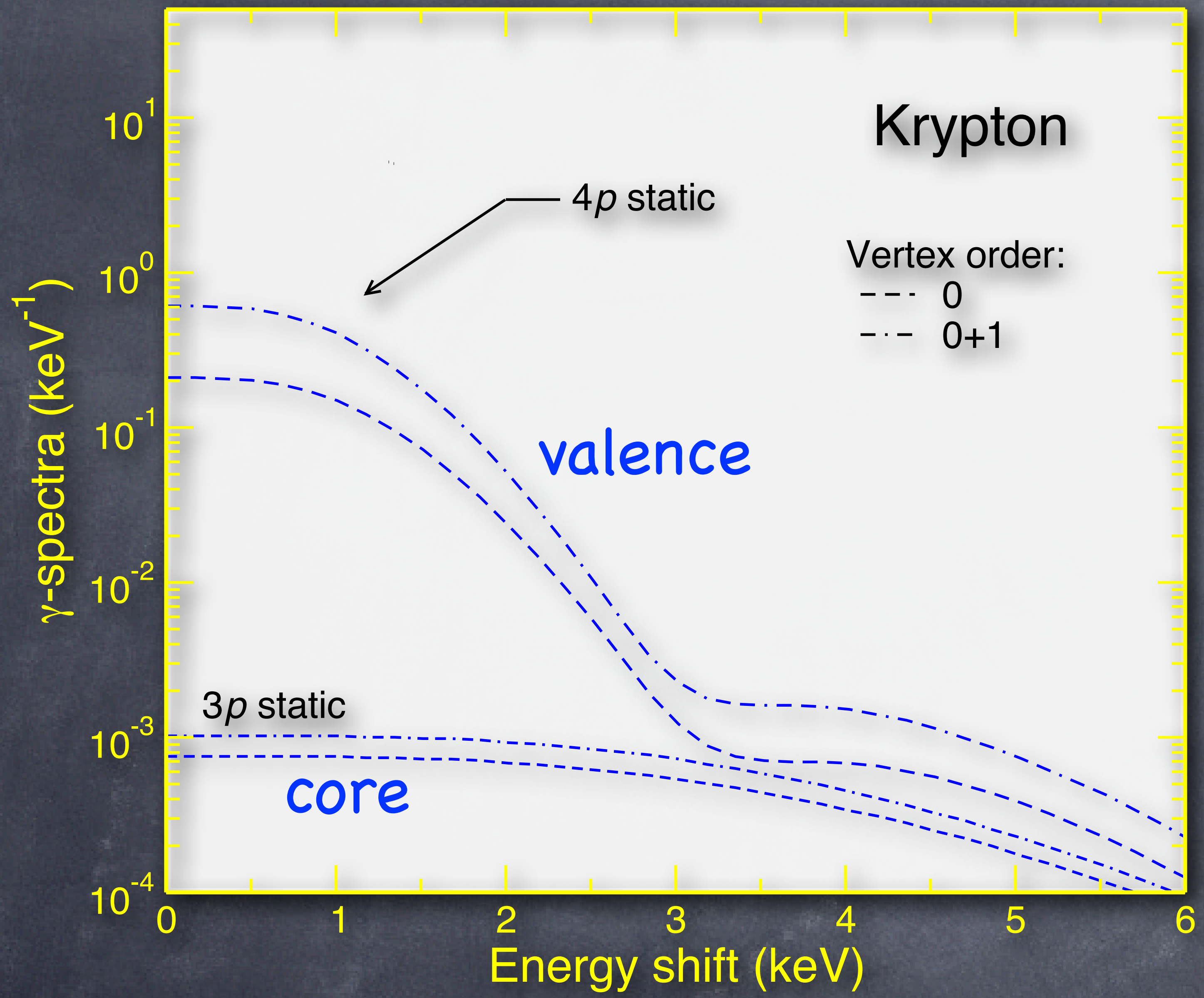
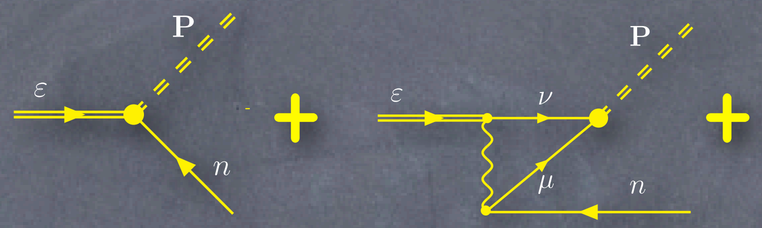
Effect of correlations: (1) VERTEX



VERTEX ENHANCEMENT



	$\frac{(0+1)}{(0)}$
Valence:	2.5
Core:	1.30

$$\lambda_{\text{ann.}} \propto \int w(\varepsilon) d\varepsilon$$

electron-positron attraction
=> increased annihilation probability

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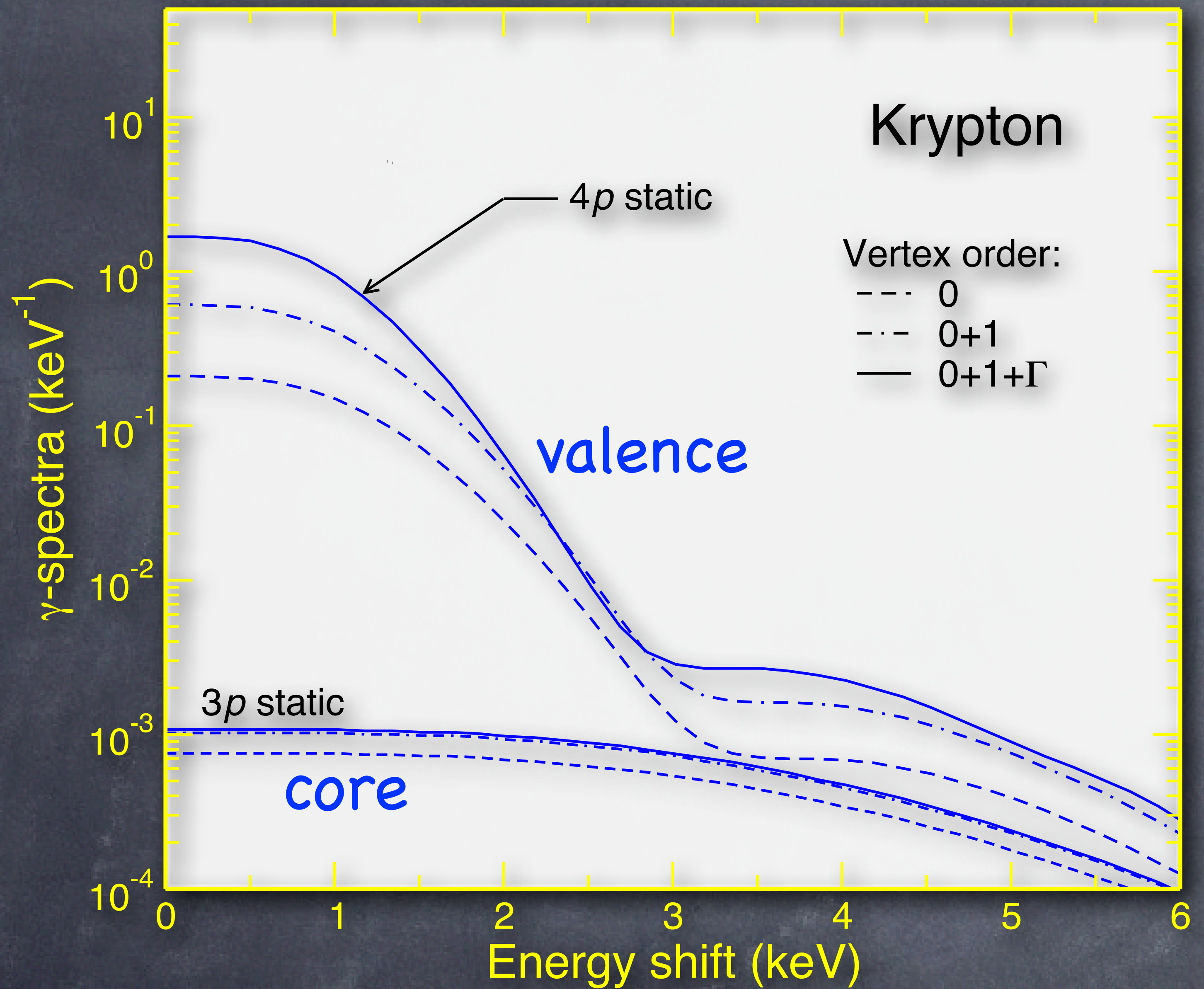
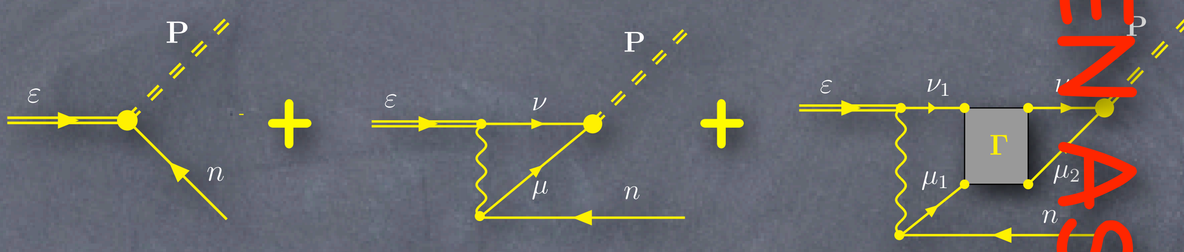
Effect of correlations: (1) VERTEX: atoms



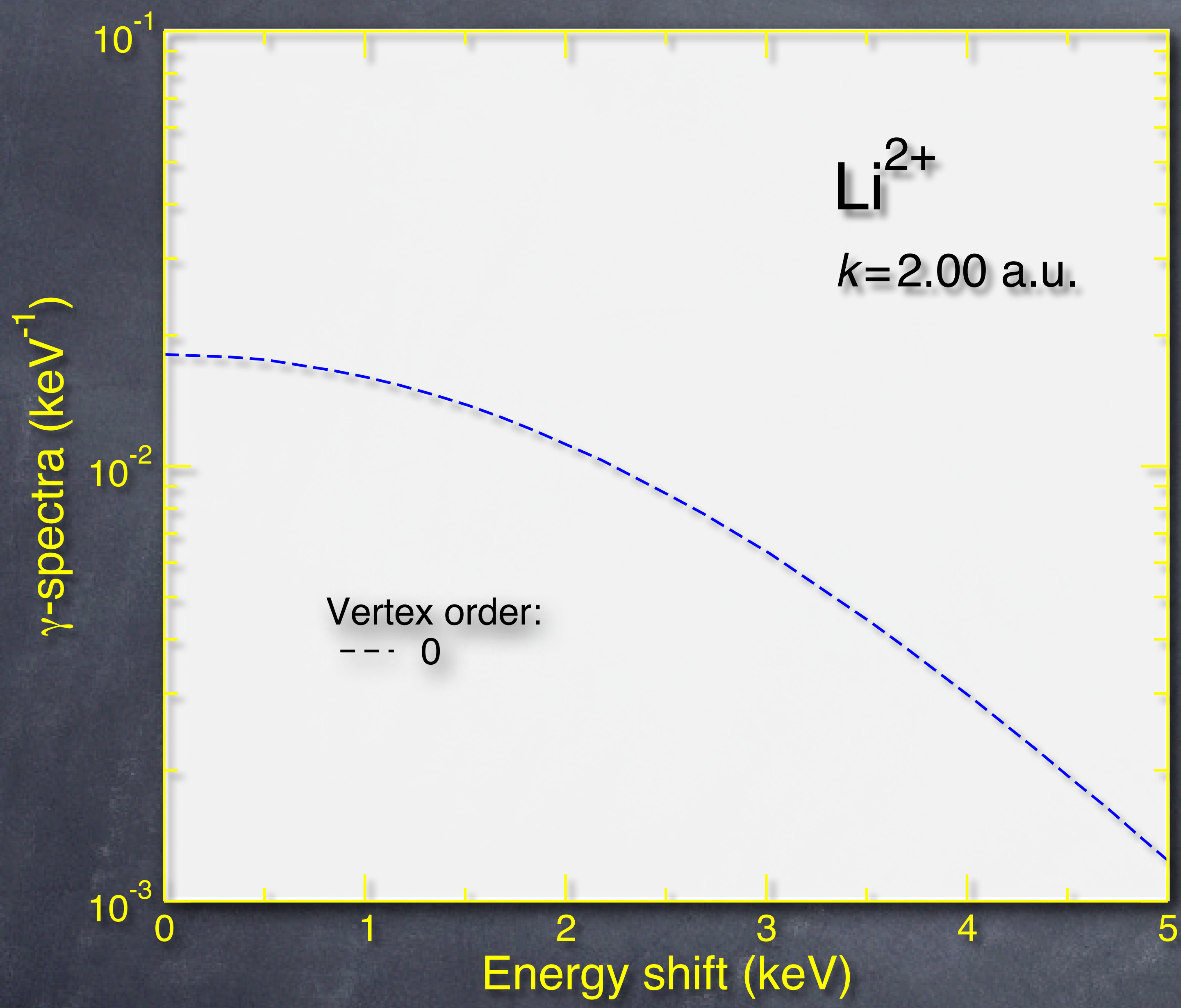
VERTEX ENHANCEMENT



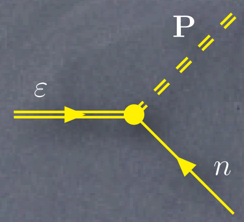
	$\frac{(0+1)}{(0)}$	$\frac{(0+1+\Gamma)}{(0)}$
Valence:	2.5	5.75
Core:	1.30	1.37

Γ-block correction:
significant for valence, small for core

Effect of correlations: (1) VERTEX: Positive ions

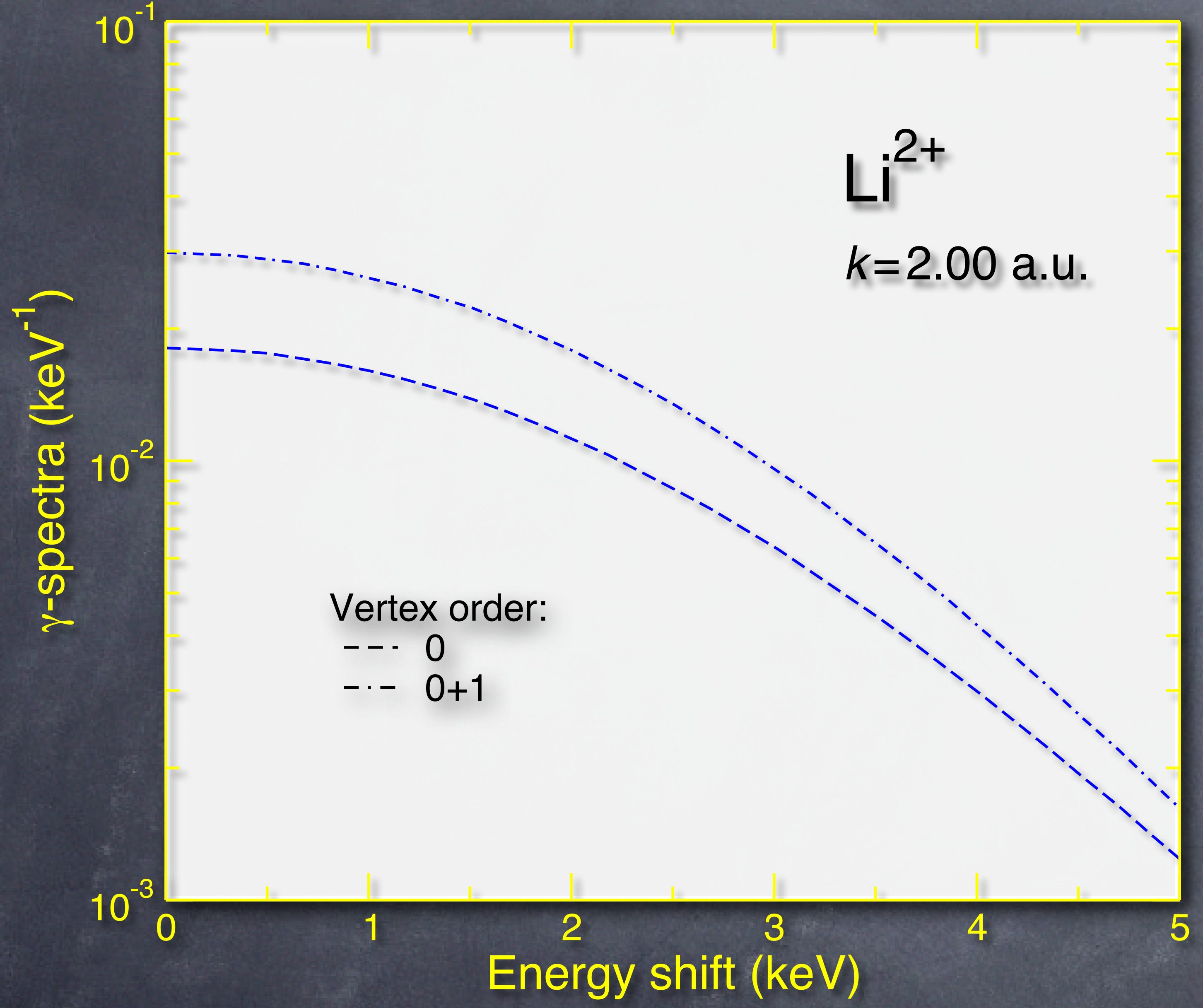


VERTEX ENHANCEMENT

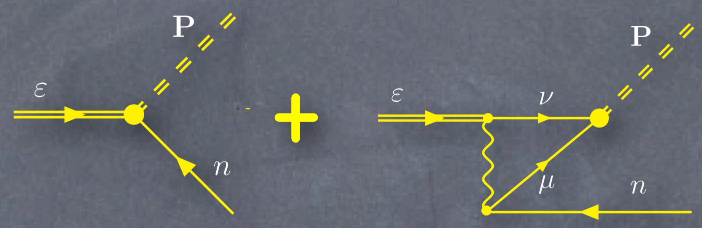


+

Effect of correlations: (1) VERTEX: Positive ions

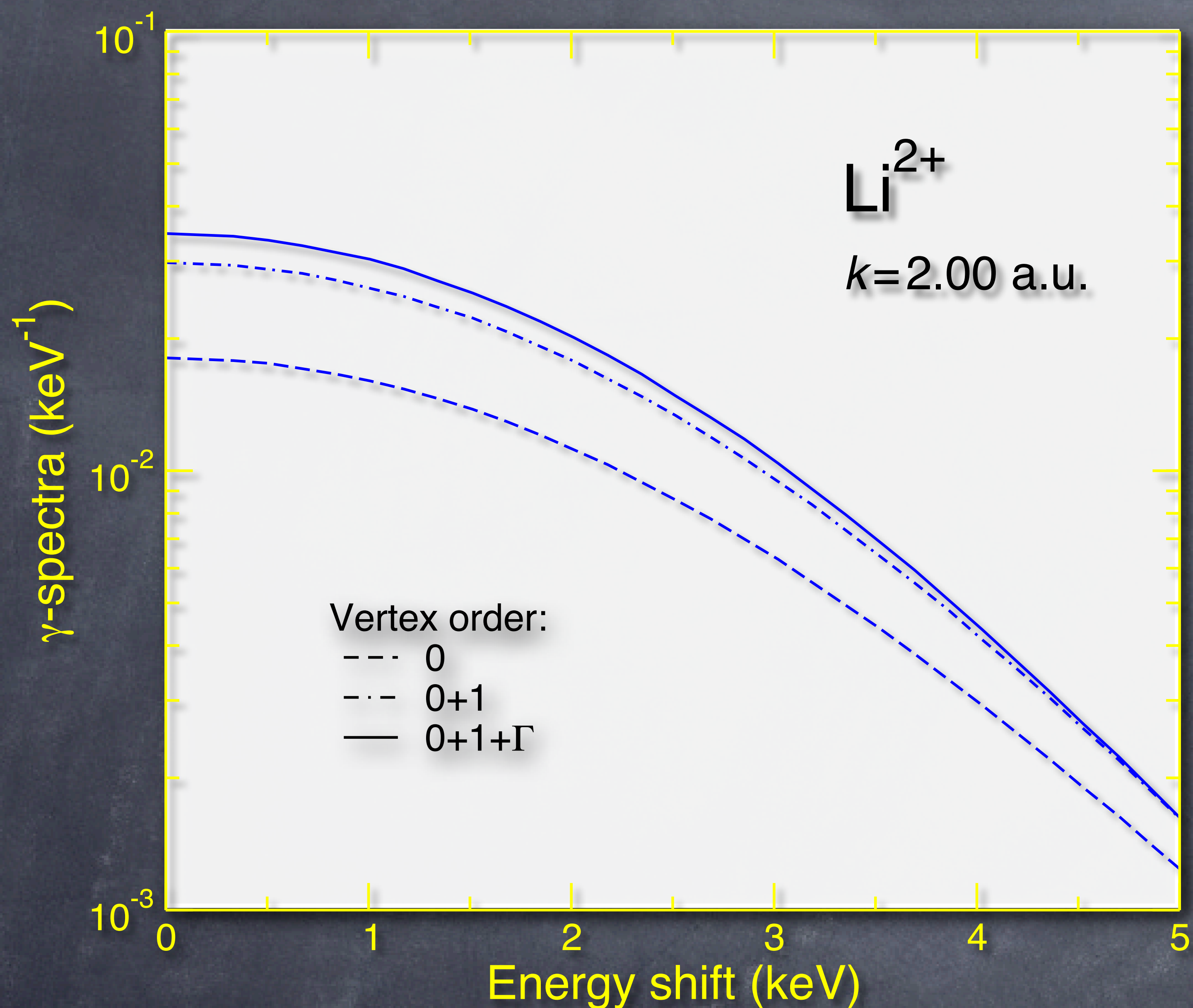


VERTEX ENHANCEMENT

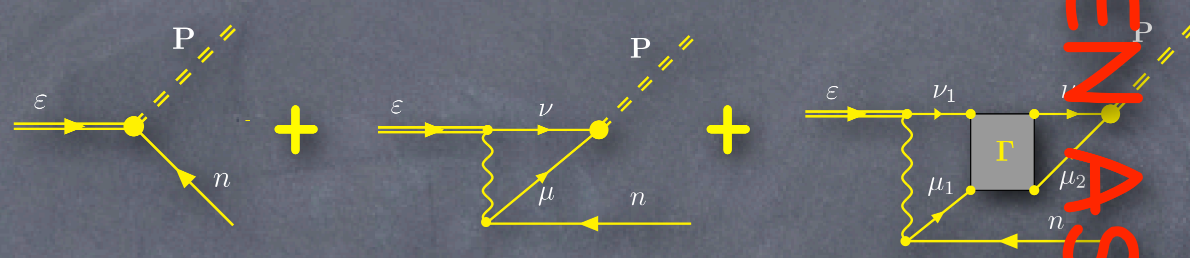


	(0+1)/(0)
1s:	1.66

Effect of correlations: (1) VERTEX: Positive ions



VERTEX ENHANCEMENT



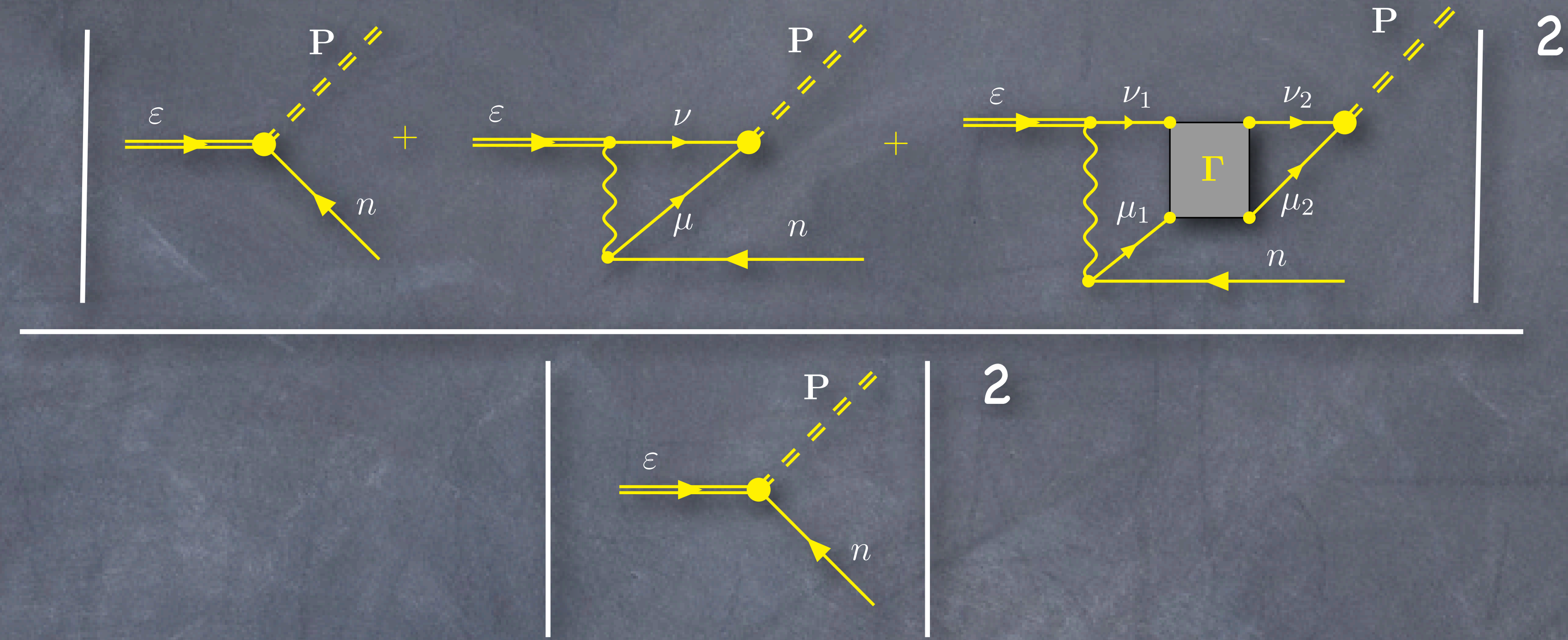
	$\frac{(0+1)}{(0)}$	$\frac{(0+1+\Gamma)}{(0)}$
1s:	1.66	1.93

H-like ions: similar behaviour to core electrons

Vertex enhancement factors

Enhancement factor:

$\bar{\gamma}_{n\varepsilon} \sim$



$$w_{n\varepsilon}(\epsilon) \approx \bar{\gamma}_{n\varepsilon}\, w_{n\varepsilon}^{\mathrm{IPM}}(\epsilon)$$

If 1st order dominates: $\bar{\gamma} - 1 \propto \dfrac{V_{\mathrm{int}}}{\Delta E}$

H-like ions: $\bar{\gamma} - 1 \propto \dfrac{1}{Z-1}$

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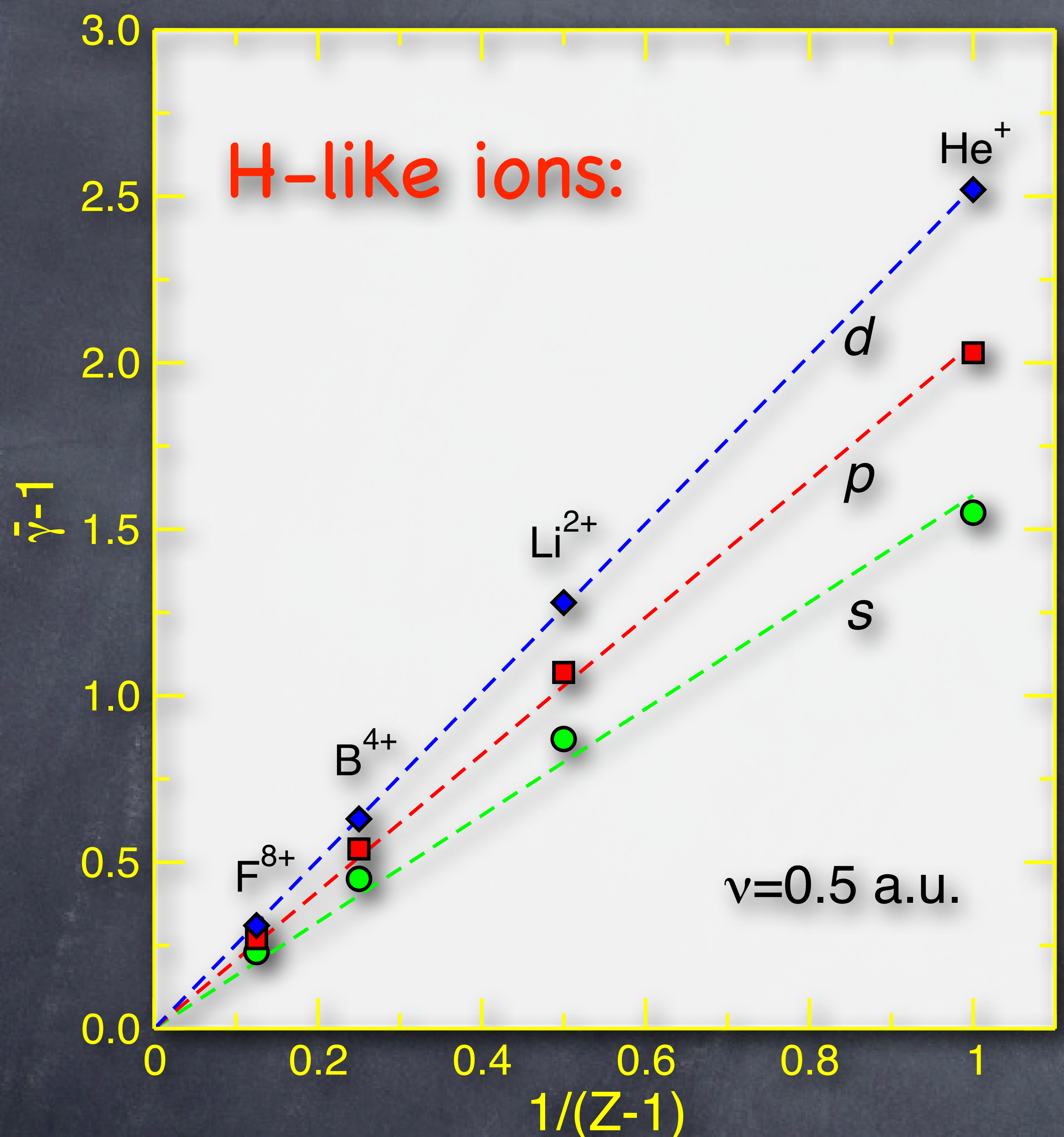
Vertex enhancement factors



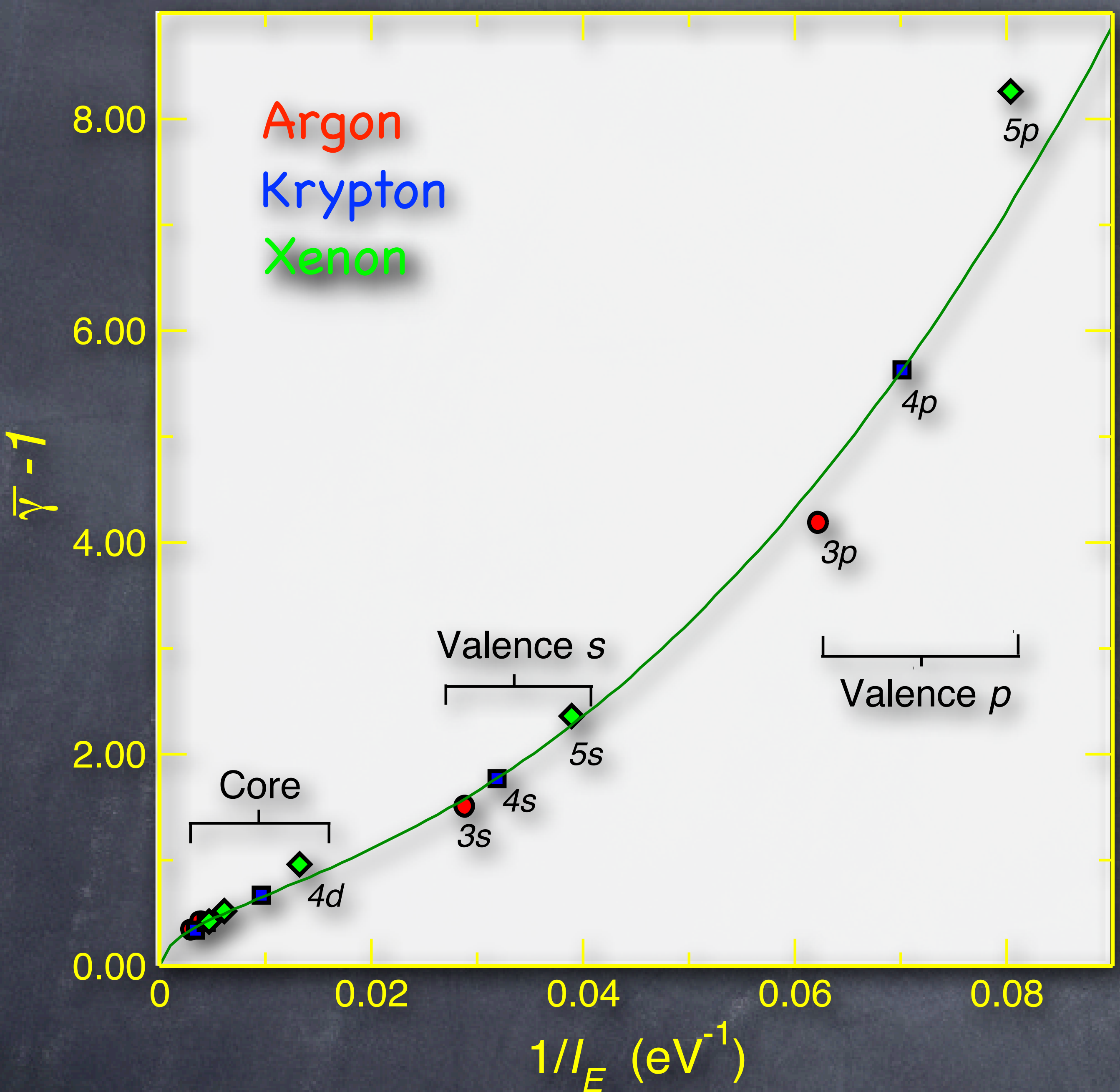
$$\bar{\gamma} - 1 \simeq X\,(Z-1)^{-1}$$

	X	
	MBT	CI-Kohn[4]
s	1.60	1.50
p	2.06	2.00
d	2.53	—

[4] Novikov et al. Phys. Rev. A, **69**, 052702 (2004)

$\bar{\gamma} - 1 \propto \dfrac{1}{Z-1}$; Ionization energy: $I \propto Z^2$; $\bar{\gamma} - 1 \propto \dfrac{1}{\sqrt{I}}$

Vertex enhancement factors

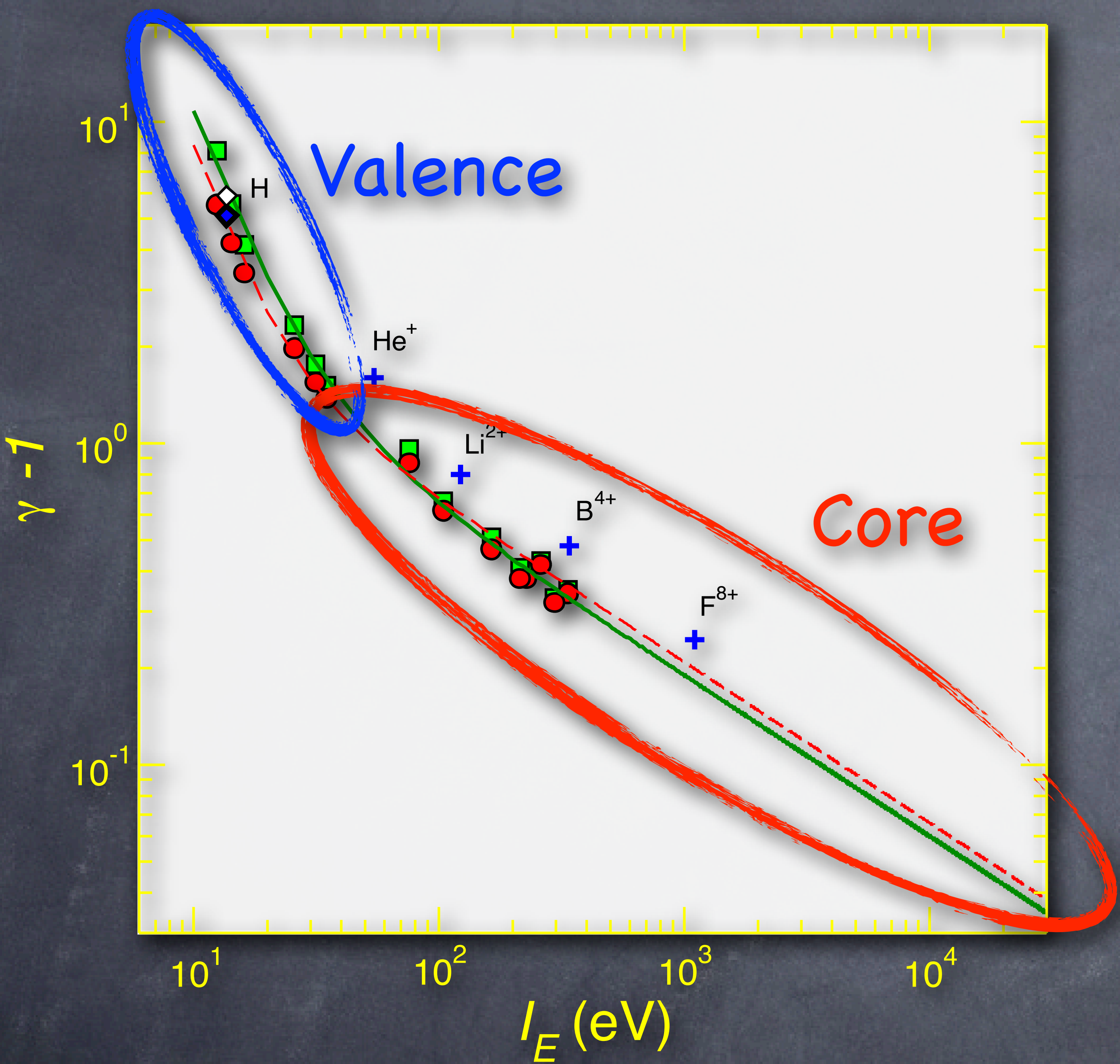


$$\bar{\gamma}_{nl} - 1 \approx \sqrt{\frac{A}{I_{nl}}} + \left(\frac{C}{I_{nl}}\right)^{\beta}$$

Core — Valence

$$A \sim 44\,\mathrm{eV}\ \ (1.6\ \mathrm{a.u.})$$
$$C \sim 20\,\mathrm{eV}\ \ (0.75\,\mathrm{a.u.})$$
$$\beta \sim 2.2$$

Vertex enhancement factors



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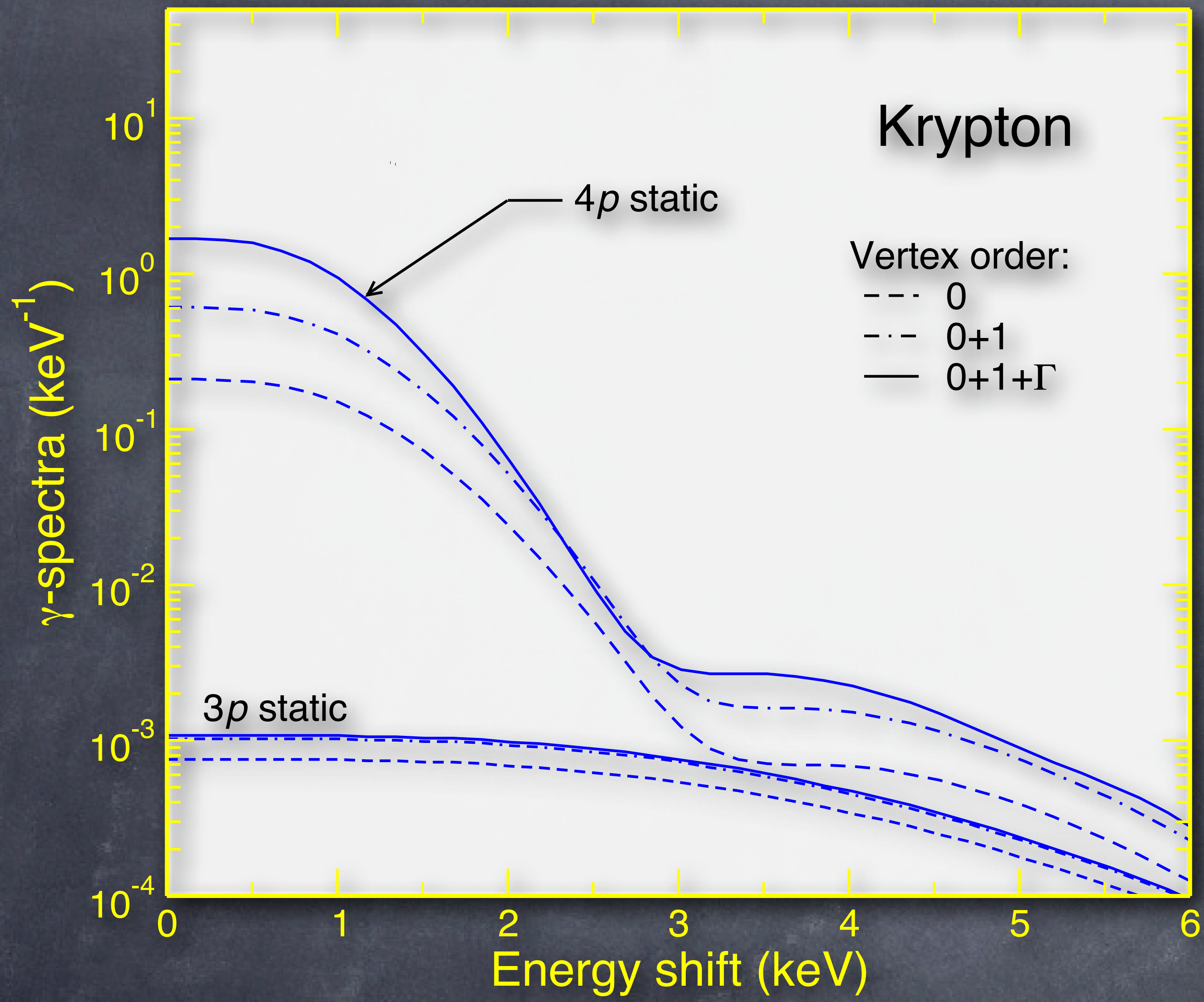
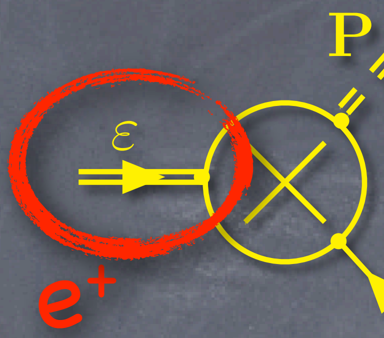
$$A \sim 44\,\mathrm{eV}\ (1.6\ \mathrm{a.u.})$$
$$C \sim 20\,\mathrm{eV}\ (0.75\ \mathrm{a.u.})$$
$$\beta \sim 2.2$$

$$w_{n\varepsilon}(\epsilon) \approx \bar{\gamma}_{n\varepsilon}\, w_{n\varepsilon}^{\mathrm{IPM}}(\epsilon)$$

Use in IPM calculations to calculate core electron spectra for:
(1) atoms across the periodic table
(2) condensed matter systems

Effect of correlations: (2) e+ WAVEFN.





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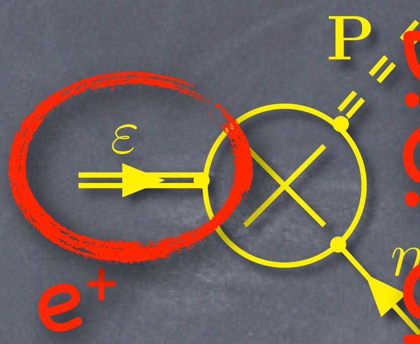
Effect of correlations: (2) e⁺ WAVEFN.



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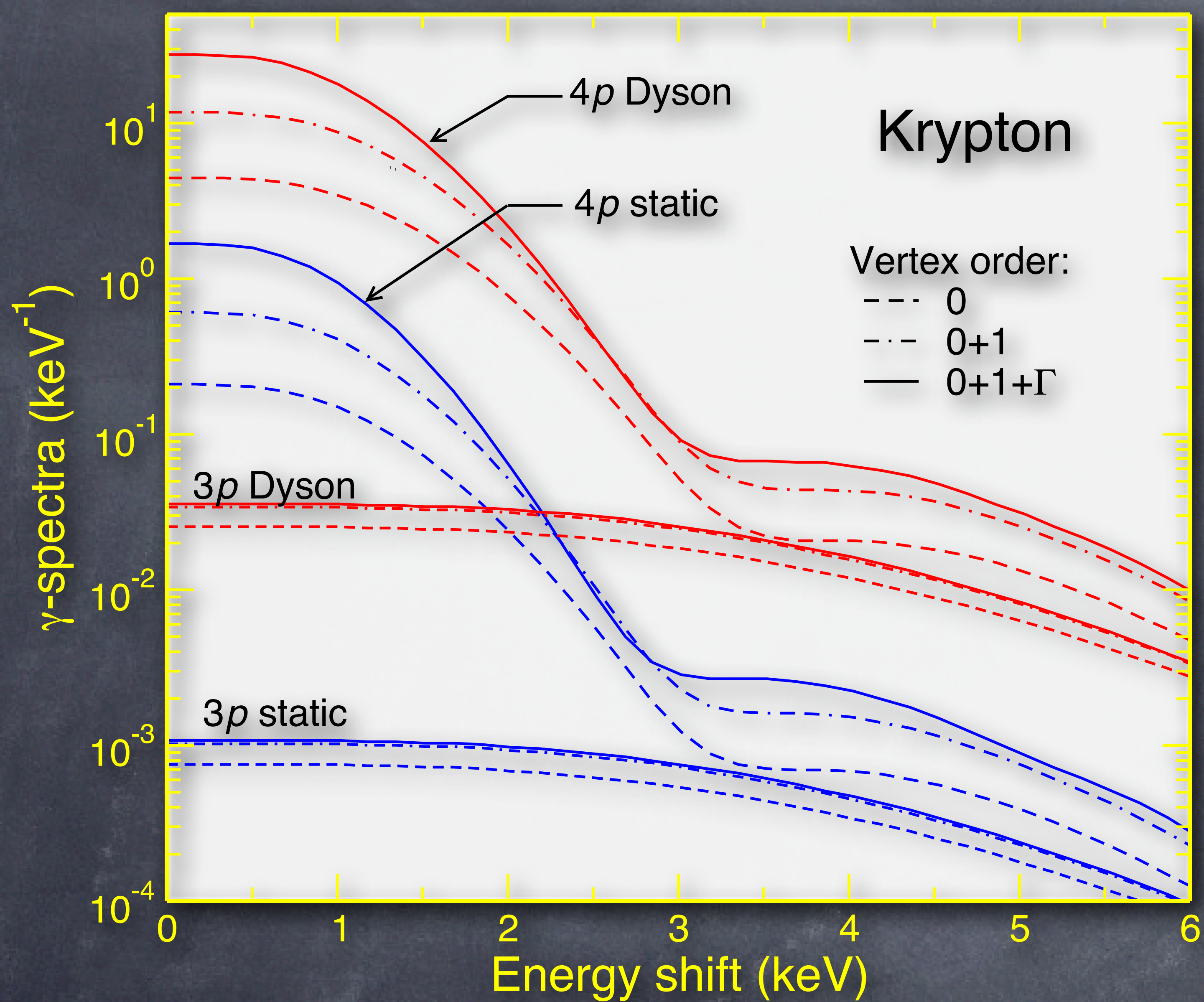
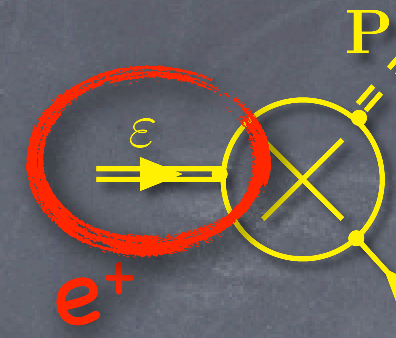


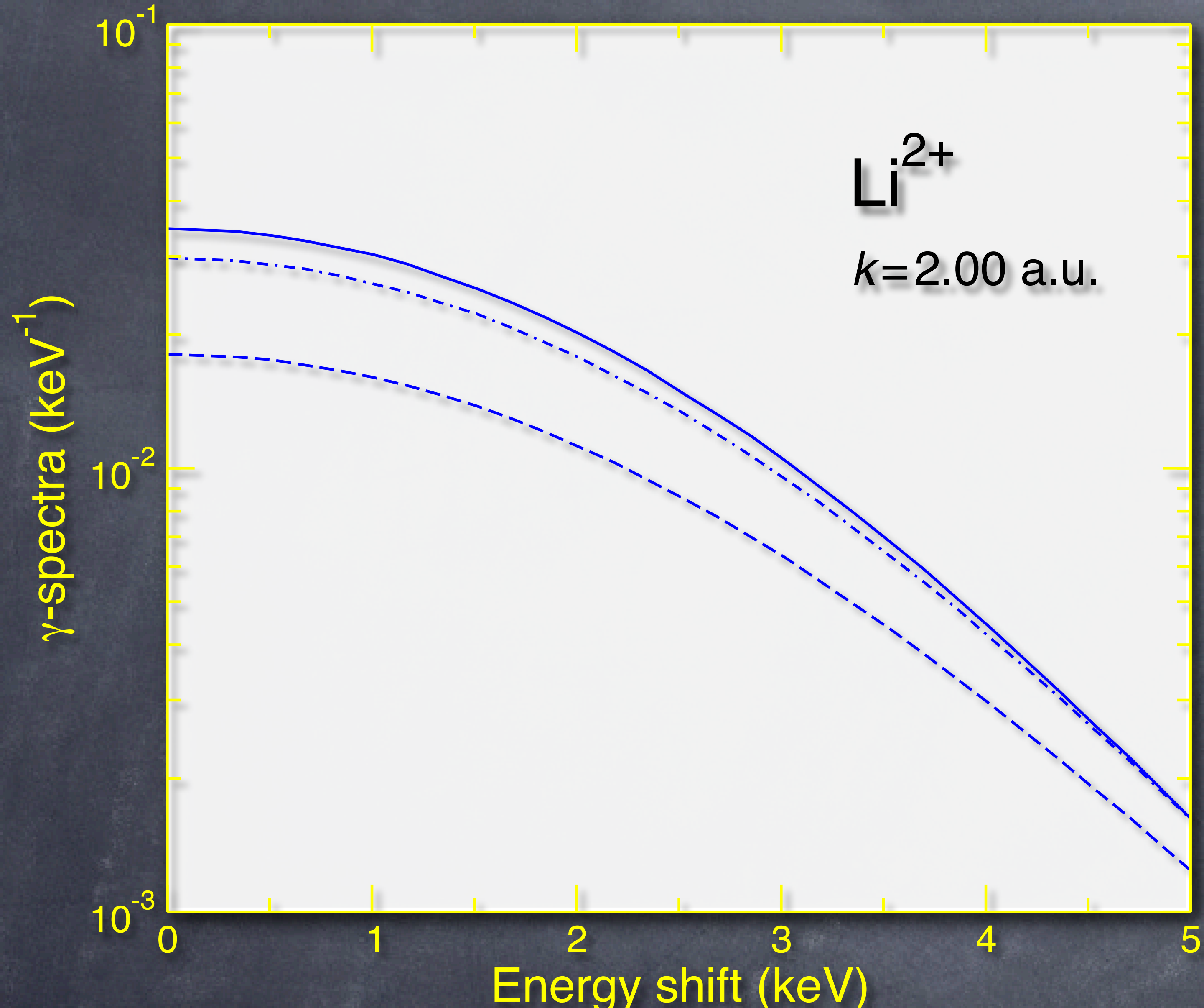
POSITRON Wavefn.

- repulsive HF potential -> attractive potential
- virtual s-states: low-energy resonance behaviour of annihilation probability [5,6]

[5] Goldanski and Sayasov, Phys. Lett., **13**, 300 (1968)
[6] Dzuba et al., Phys. Scripta T46 (1993)

Effect of correlations: (2) e⁺ WAVEFN.





POSITRON Wavefn.

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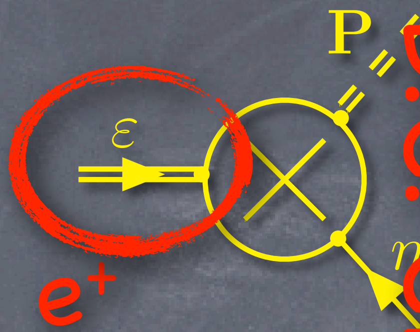
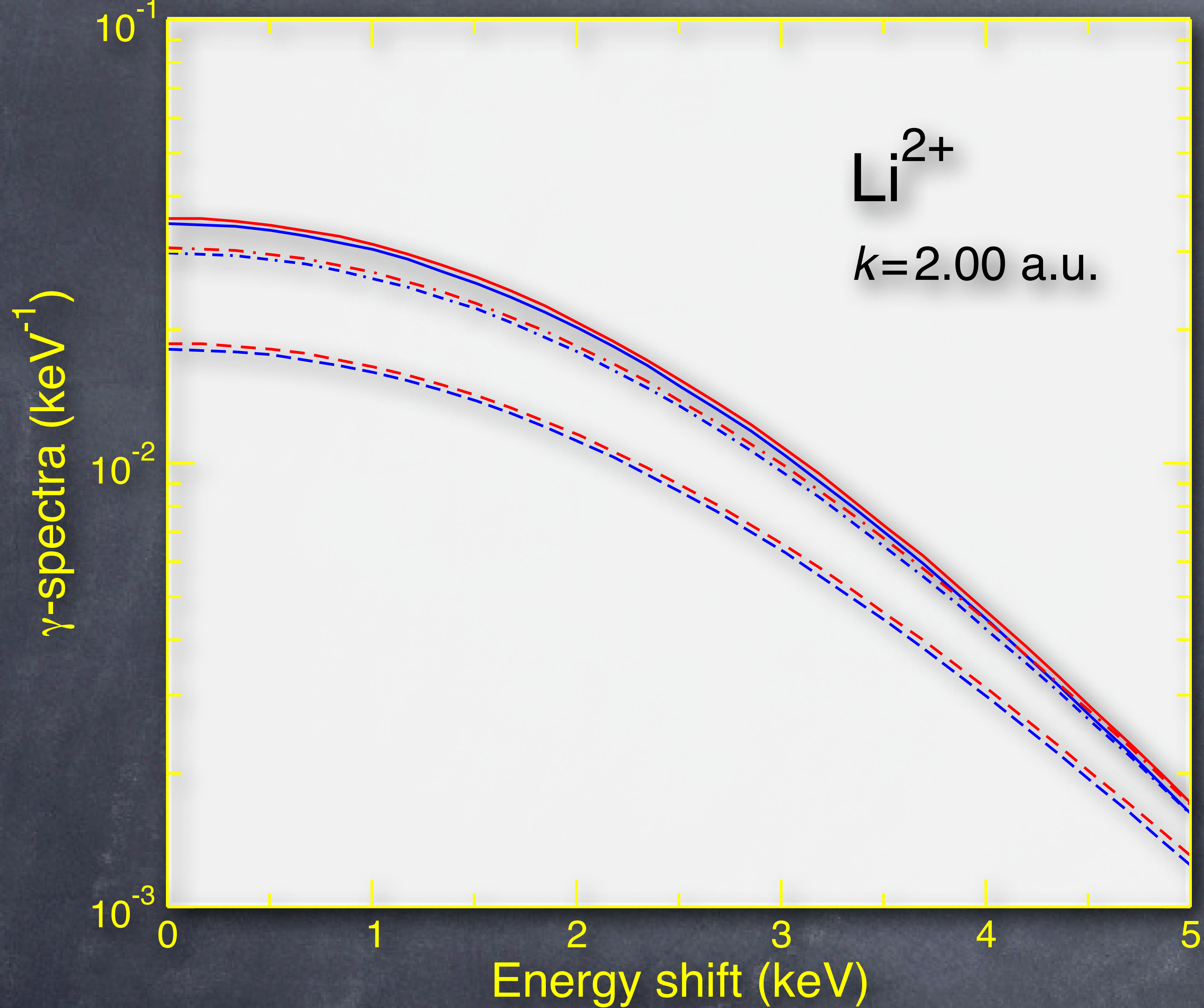
Effect of correlations: (2) e⁺ WAVEFN.





<u>POSITRON Wavefn.</u>

Nuclear repulsion dominates!

...only vertex enhancement important

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Outline

1. Many-body theory of annihilation γ-spectra
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Comparison with Expt.

Argon

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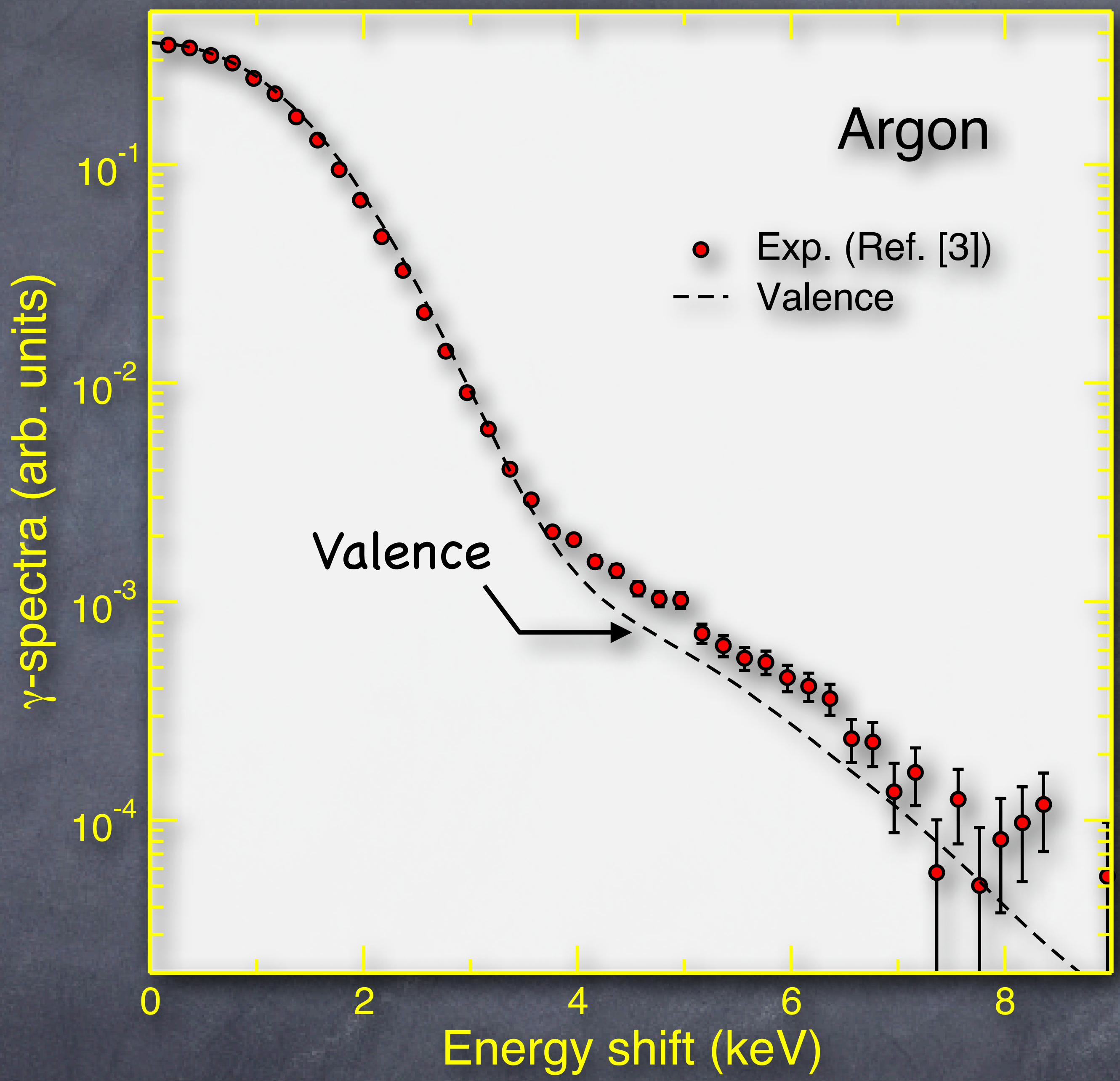
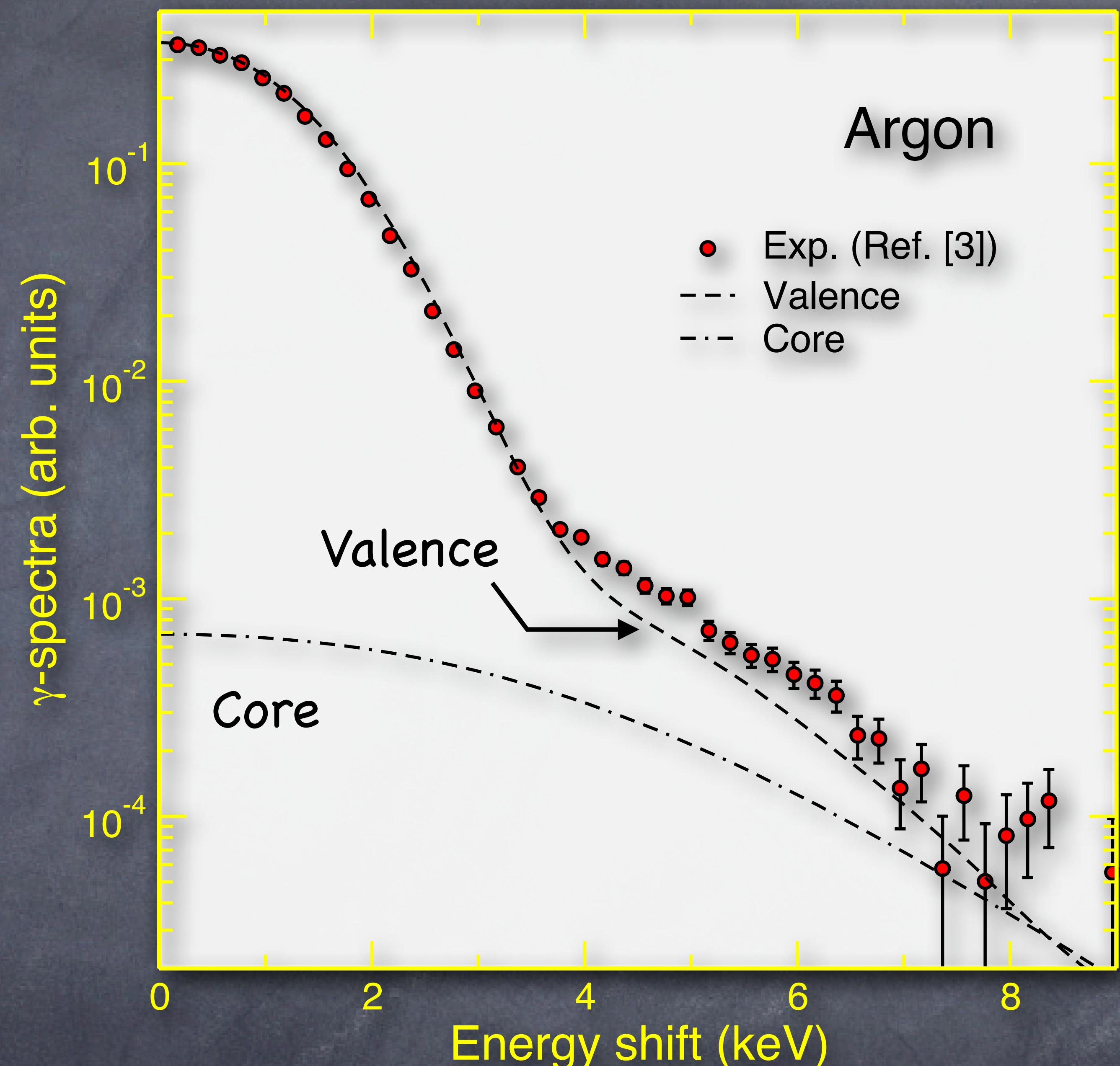


Annihilation on
valence shells insufficient!

[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Comparison with Expt.

Argon



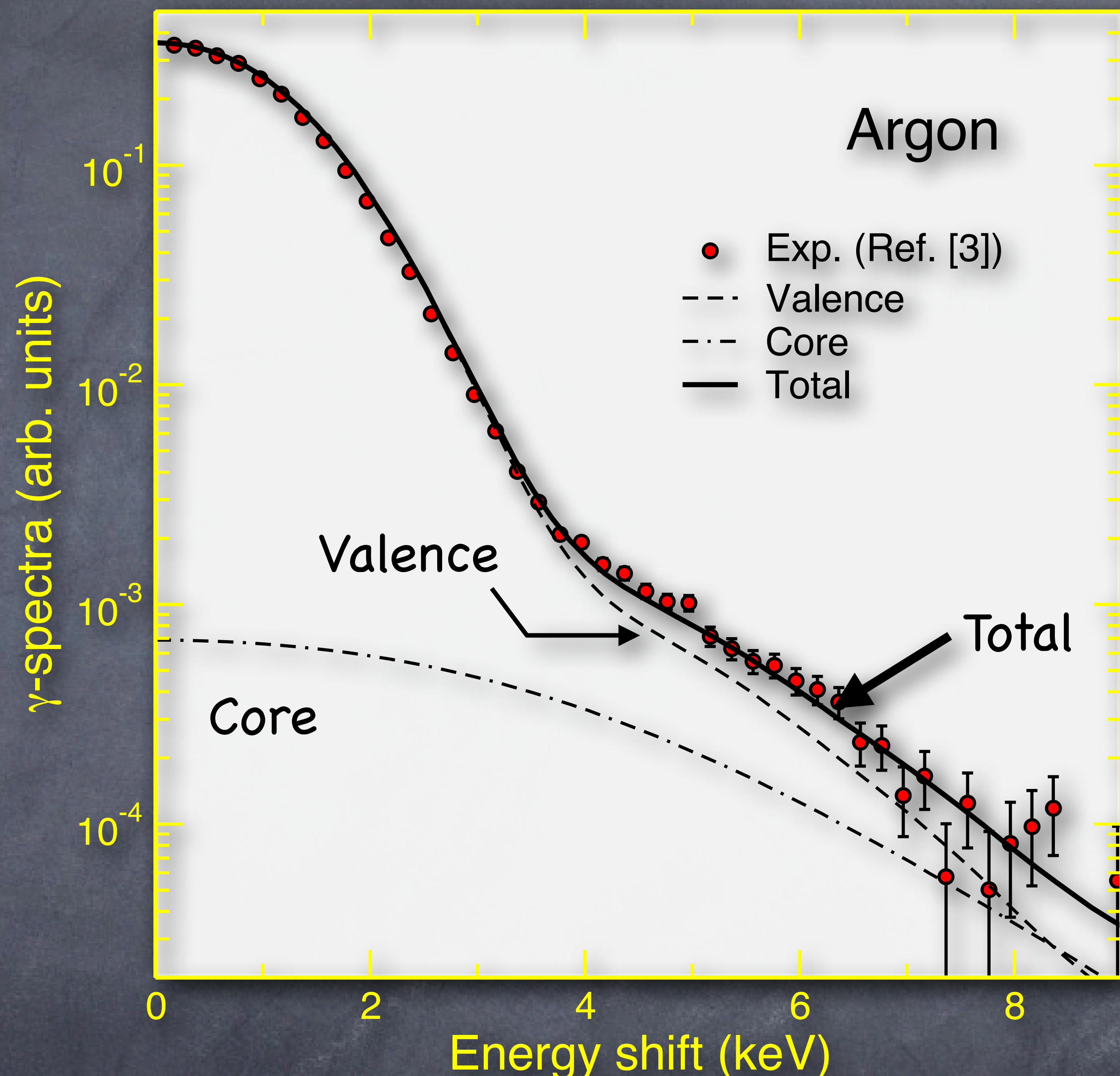
	% of tot ann. rate
Val.	99.4%
Core	0.6%

Nuc. repulsion =>
Ann. predominantly occurs on valence shells

[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Comparison with Expt.

Argon



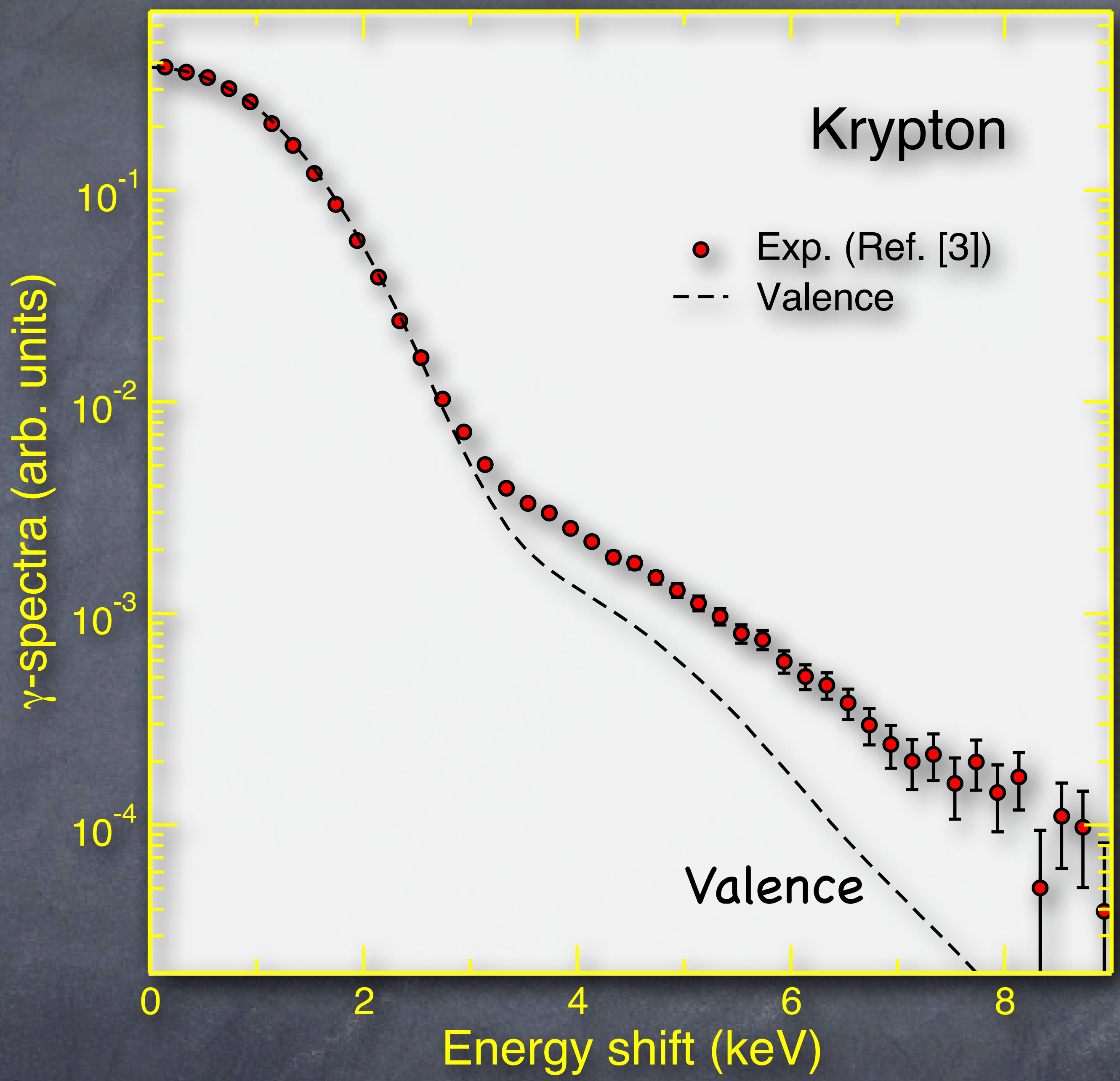
	% of tot ann. rate
Val.	99.4%
Core	0.6%

Core contribution is vital to obtain agreement with experiment!

[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Comparison with Expt.

Krypton

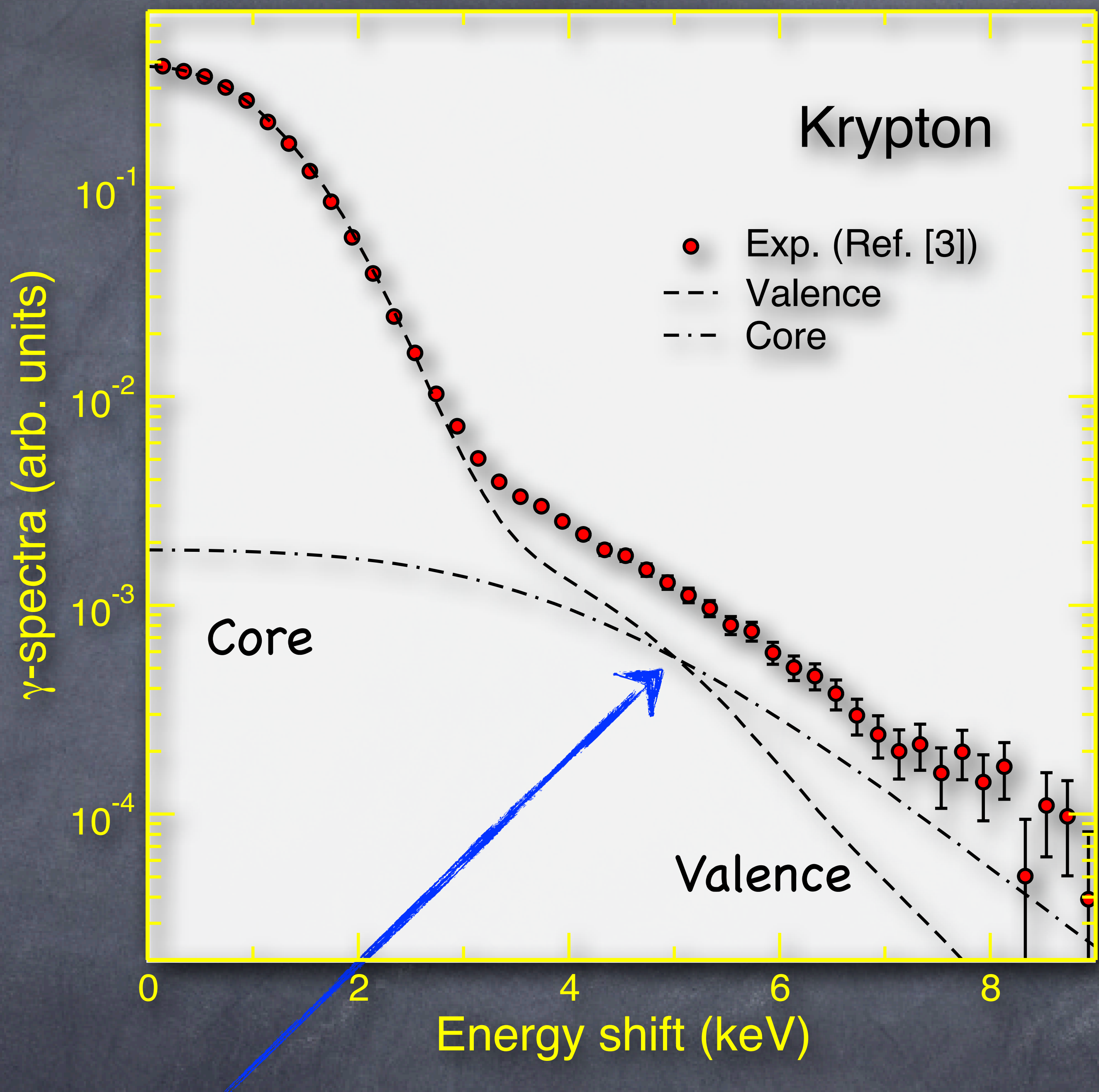


Valence shells: severely underestimate high-energy `wing'

[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Comparison with Expt.

Krypton



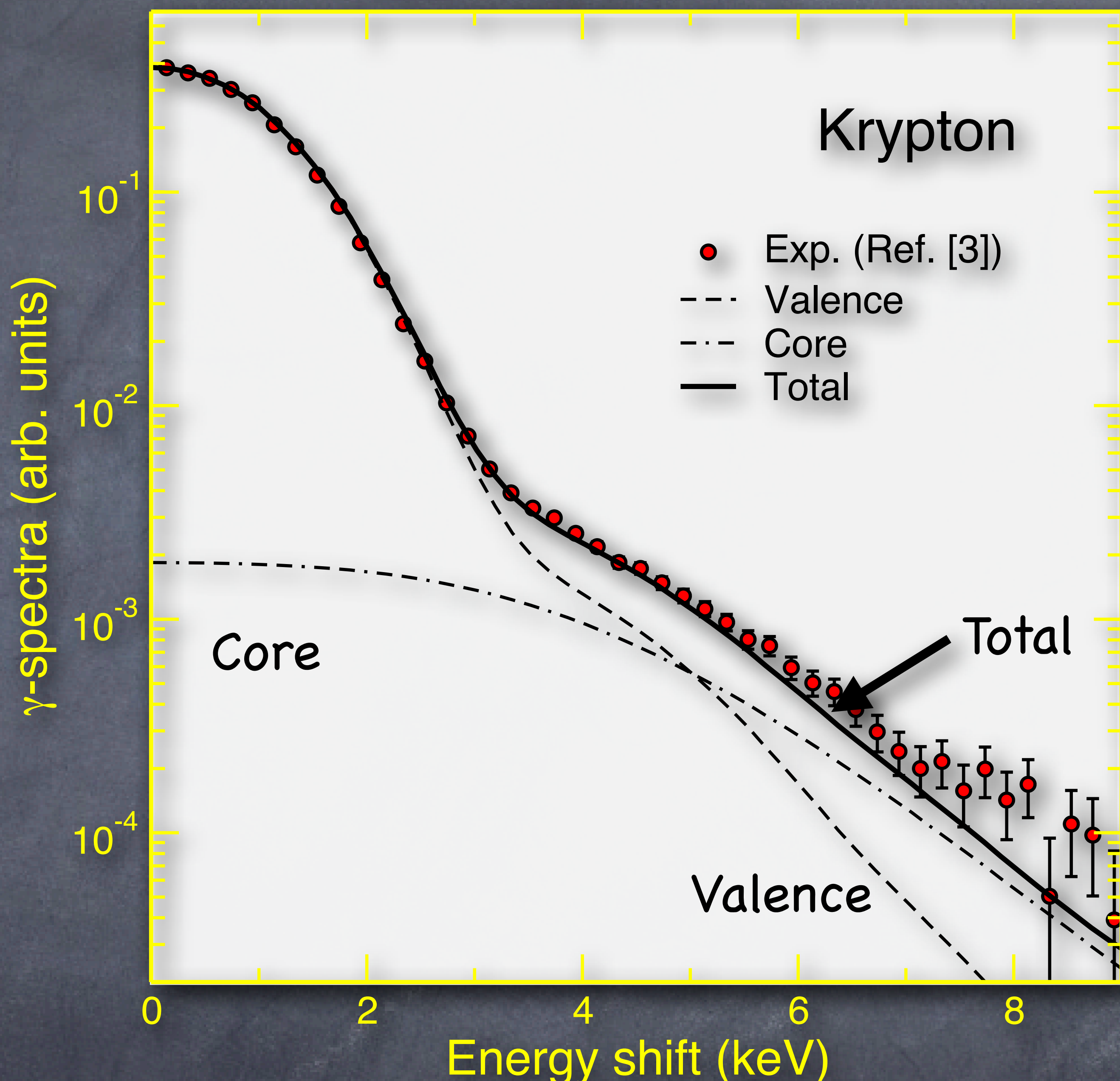
	% of tot ann. rate
Val.	98.5%
Core	1.5%

Core contribution DOMINATES above 5keV

[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Comparison with Expt.

Krypton



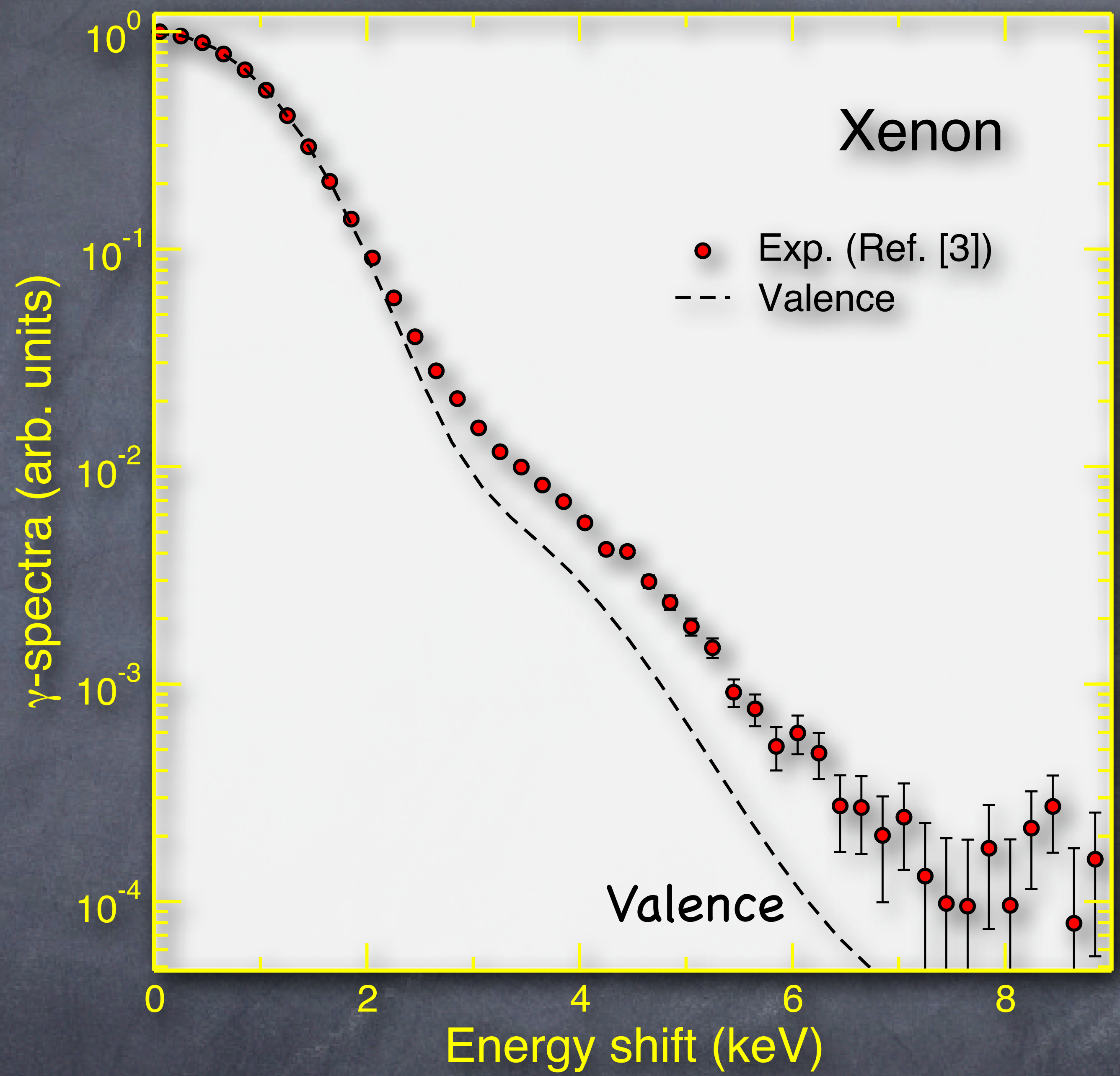
	% of tot ann. rate
Val.	98.5%
Core	1.5%

Adding core contribution results in good agreement!
(discrepancy above 6 keV...?)

[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

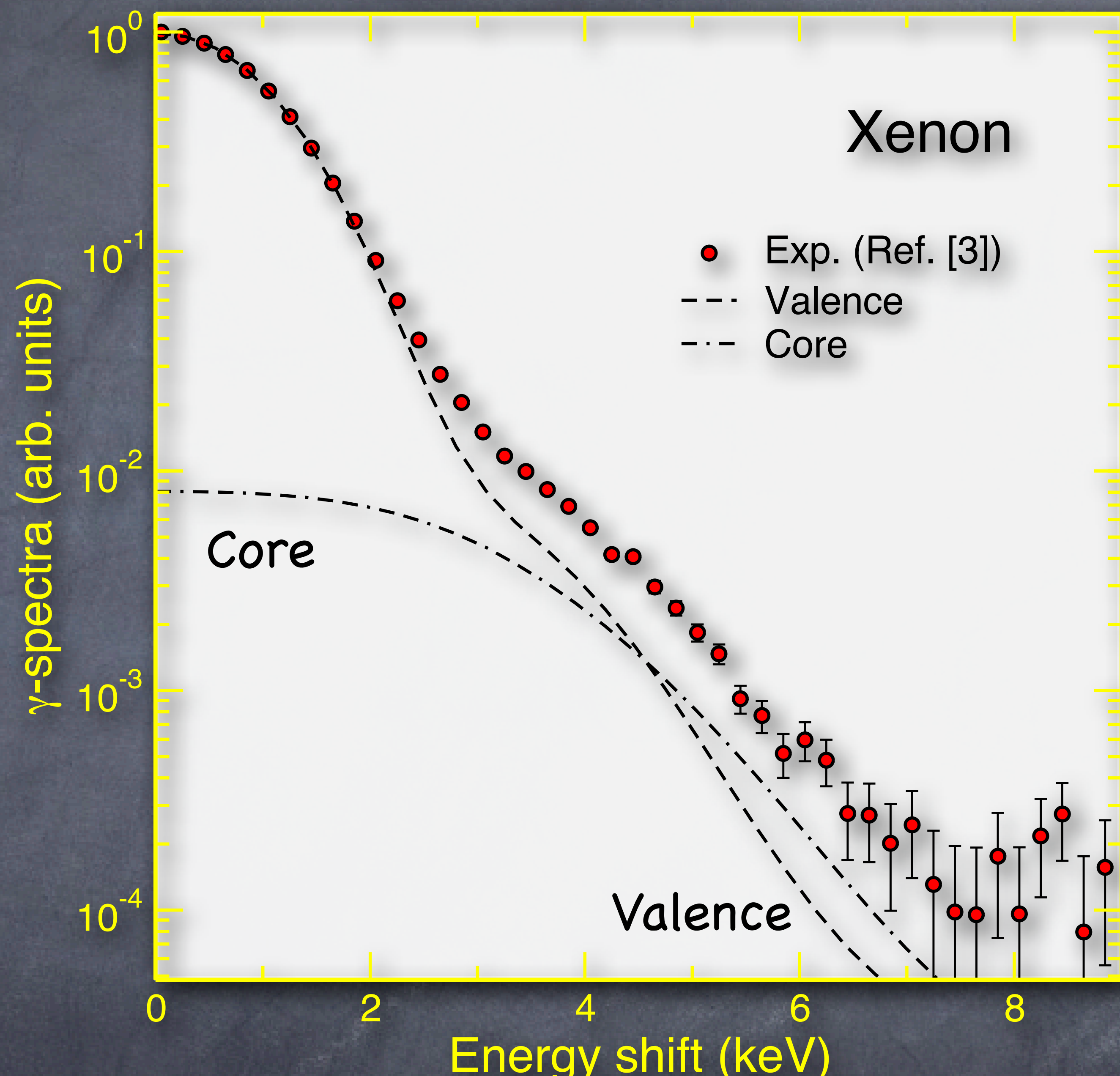
Comparison with Expt.

Xenon



[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Comparison with Expt.



Xenon

	% of tot ann. rate
Val.	97.8%
Core	2.2%

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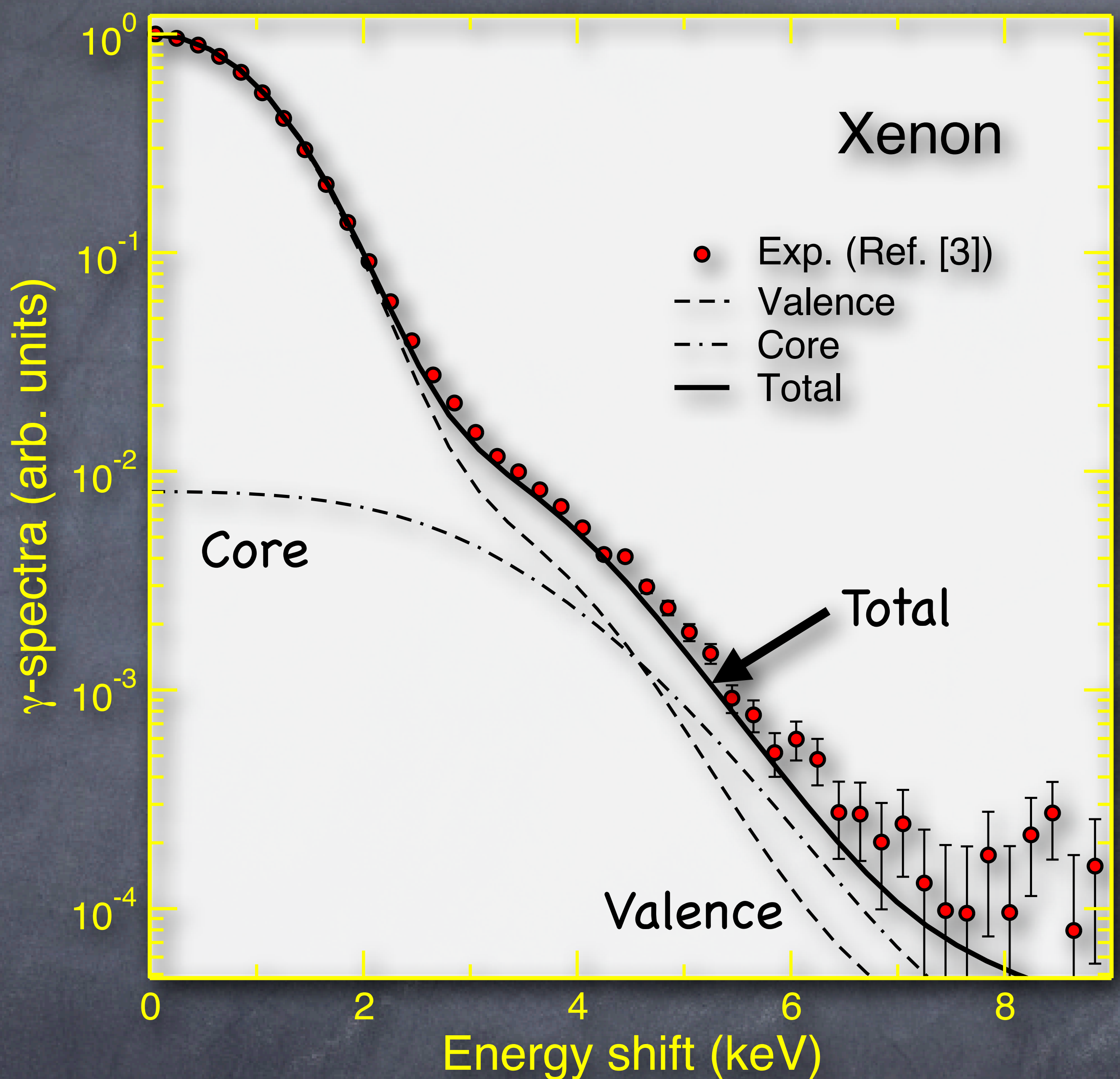
[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Comparison with Expt.

Xenon



	% of tot ann. rate
Val.	97.8%
Core	2.2%

Ar->Kr->Xe

core annihilation progressively more important!

[3] Iwata et al., Phys. Rev. Lett **79** 39 (1997)

Conclusions

- Ab initio MBT calculations of γ-spectra for core and valence shells of many-electron atoms + positive ions.
- Positron-atom and (short-range) electron-positron correlations are stronger for the valence electrons, but are still significant for the core! (and for H-like ions of Z<10).
- `True' vertex enhancement factors have been calculated and follow a simple scaling with the electron ionization energy.
- Despite the small annihilation rates (owing to nuclear repulsion), the core contributes significantly to the spectrum at large Doppler shifts.

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