

Two-photon emission from few-electron heavy ions: Angle- and polarization-resolved studies

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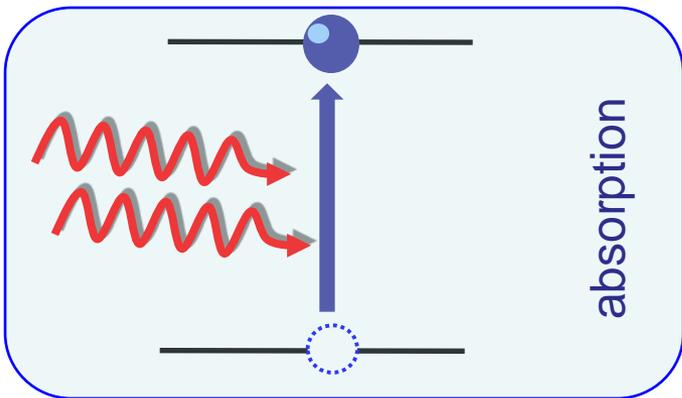
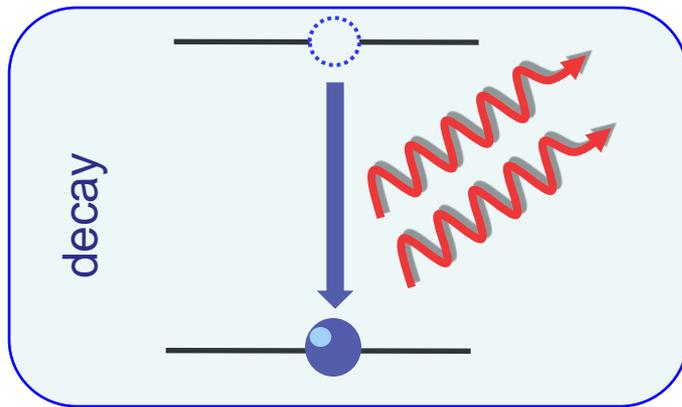


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Two-photon atomic transitions

- In 1930 in her PhD thesis Maria Göppert-Mayer suggested that bound-bound transitions can undergo under simultaneous absorption/emission of two correlated photons.



Über Elementarakte mit zwei Quantensprüngen

Von Maria Göppert-Mayer

(Göttinger Dissertation)

(Mit 5 Figuren)

Einleitung

Der erste Teil dieser Arbeit beschäftigt sich mit dem Zusammenwirken zweier Lichtquanten in einem Elementarakt. Mit Hilfe der Diracschen Dispersionstheorie¹⁾ wird die Wahrscheinlichkeit eines dem Ramaneffekt analogen Prozesses, nämlich der Simultanemission zweier Lichtquanten, berechnet. Es zeigt sich, daß eine Wahrscheinlichkeit dafür besteht, daß ein angeregtes Atom seine Anregungsenergie in zwei Licht-

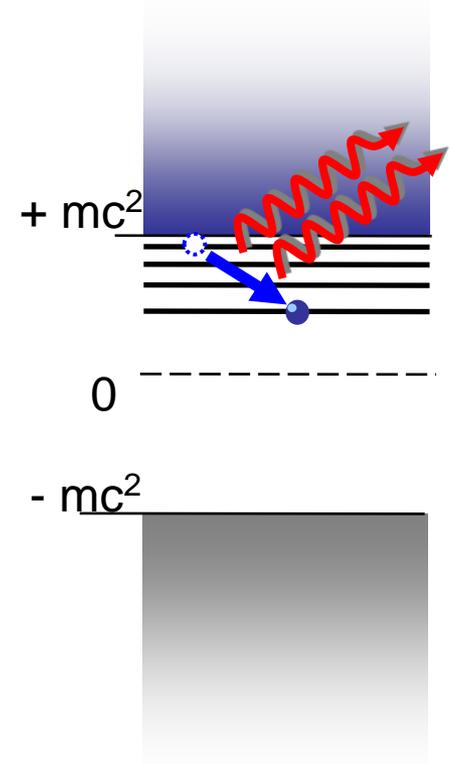
- For many years two-photon decay/excitation of atoms and ions has attracted much of experimental and theoretical interest.

Two-photon atomic transitions

- Analysis of the two-photon decay requires knowledge about complete spectrum of the:

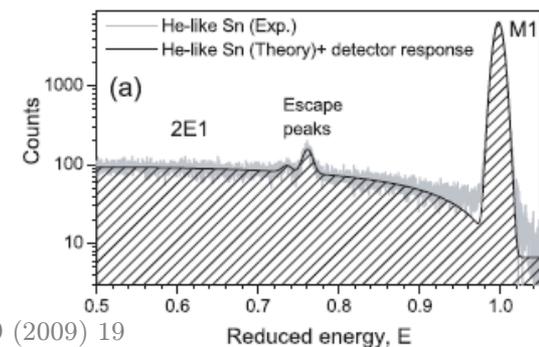
$$\tilde{M}_{fi} \propto \sum_{\nu} \frac{\langle \psi_f | \alpha \boldsymbol{\varepsilon}_2 e^{-i\mathbf{k}r_2} | \psi_{\nu} \rangle \langle \psi_{\nu} | \alpha \boldsymbol{\varepsilon}_1 e^{-i\mathbf{k}r_1} | \psi_i \rangle}{E_{\nu} - E_i + \hbar\omega_1}$$

- ◆ The summation in the second-order transition amplitude includes a summation over the discrete part of the spectrum as well as an integration over the positive and negative-energy continuum.



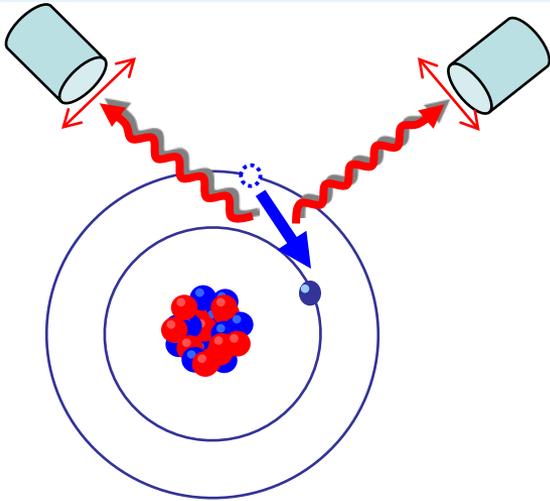
- A large number of studies have been performed over the last decades to investigate:

- ▶ Total decay rates
- ▶ Energy (spectral) distributions



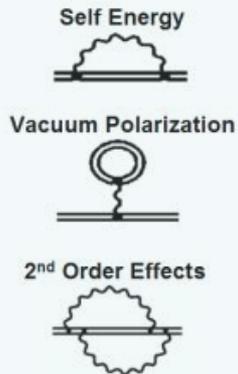
A. Kumar *et al.*, EPJ ST 169 (2009) 19

Two-photon atomic transitions: Novel studies



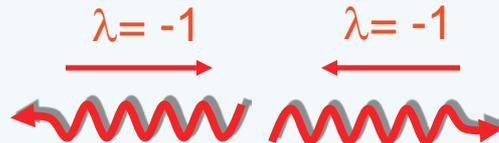
- ▶ Most recent years interest arises to study angular and polarization properties of emitted photons.
- ▶ A number of interesting applications have been proposed for these studies in high-Z domain.

- ▶ Detailed analysis of relativistic and QED effects.

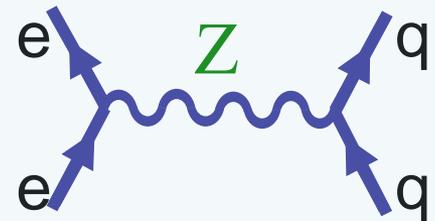


- ▶ Spin entanglement in relativistic regime, test of Bell's inequality

$$\frac{|\uparrow\uparrow\rangle + |\downarrow\downarrow\rangle}{\sqrt{2}}$$



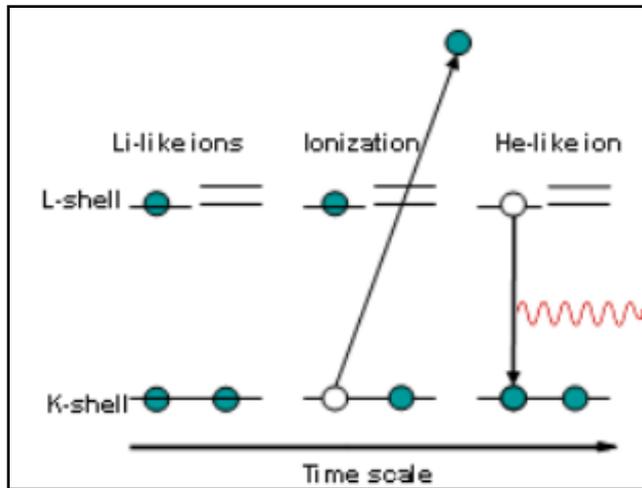
- ▶ Parity violation in heavy atomic systems



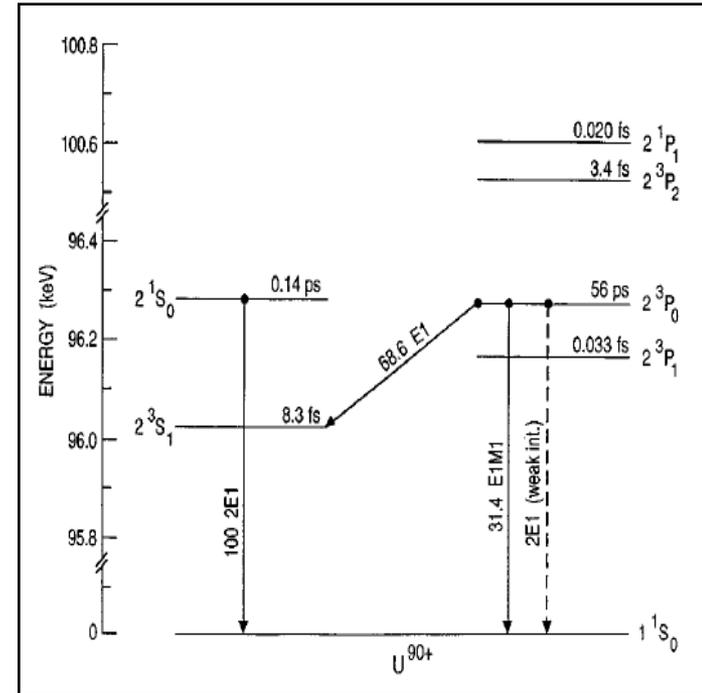
Helium-like heavy ions

► In high-Z domain two-photon studies are most likely to be performed with the helium-like ions:

- ⊕ There are metastable states even for high-Z
- ⊕ Can be efficiently “prepared” in ionic collisions
- ⊕ Good candidates for PNC studies



J. Rzakiewicz et al, PRA 74 (2006) 012511



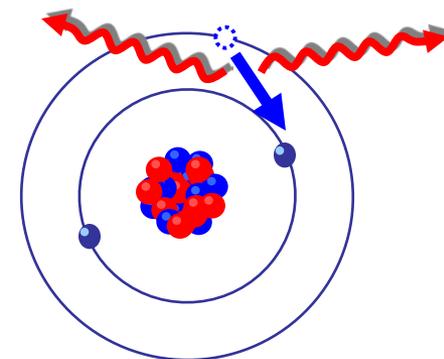
R. Dunford, PRA 54 (1996) 3820

- ◆ Request for theoretical description of two-photon transitions in helium-like heavy ions with the special emphasis on angular and polarization properties.

A little bit of theory...

- ▶ To describe two-photon transitions in helium-like heavy ions we make use of the independent particle model

$$\Psi_{JM_J}(\mathbf{r}_1, \mathbf{r}_2) = N \cdot \sum_{\mu_a \mu_b} (j_a \mu_a j_b \mu_b | JM_J) \begin{vmatrix} \psi_{n_a j_a \mu_a}(\mathbf{r}_1) & \psi_{n_b j_b \mu_b}(\mathbf{r}_1) \\ \psi_{n_a j_a \mu_a}(\mathbf{r}_2) & \psi_{n_b j_b \mu_b}(\mathbf{r}_2) \end{vmatrix}$$



$$\tilde{M}_{fi} \propto \sum_{\nu} \frac{\langle \psi_f | \mathbf{a} \cdot \boldsymbol{\varepsilon}_{\lambda_1} e^{-i\mathbf{k}r_1} | \psi_{\nu} \rangle \langle \psi_{\nu} | \mathbf{a} \cdot \boldsymbol{\varepsilon}_{\lambda_2} e^{-i\mathbf{k}r_2} | \psi_i \rangle}{E_{\nu} - E_i + \hbar\omega_2}$$

- ▶ ... which allows to express two-electron matrix elements in terms of single-electron ones.

● What are the $\psi_{nj\mu}(\mathbf{r})$ functions? We have used few approximations:

- ▶ Dirac-Coulomb functions with effective charge
- ▶ Dirac-Fock solutions
- ▶ Kohn-Sham solutions



A little bit of theory...

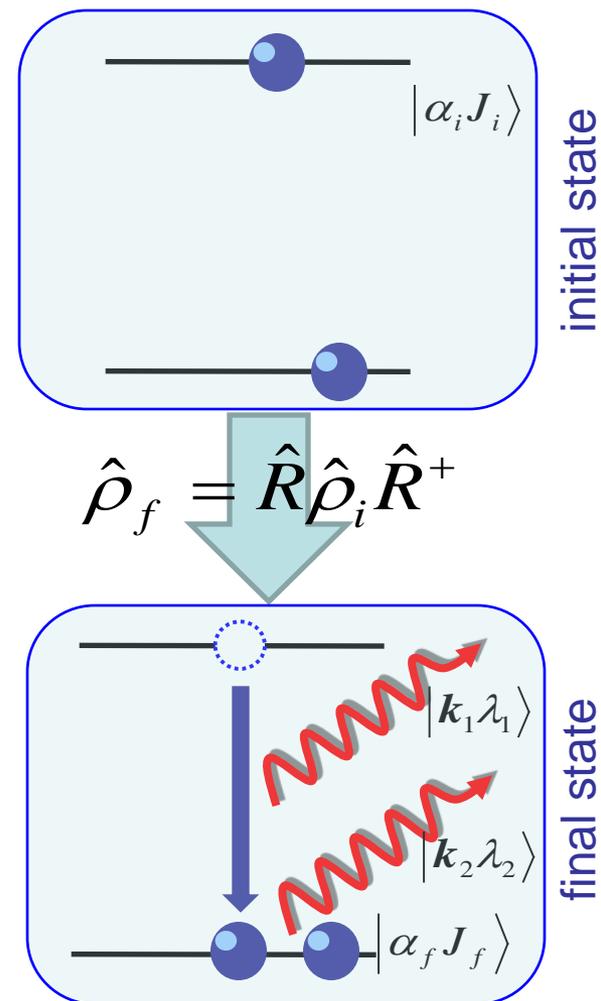
- ▶ When (two-electron) second-order transition amplitudes are combined within the framework of the density matrix theory one gets simple access to various properties of two-photon transitions.

$$\tilde{M}_{fi} \propto \sum_{\nu} \frac{\langle \psi_f | \alpha \epsilon_{\lambda_1} e^{-ikr_1} | \psi_{\nu} \rangle \langle \psi_{\nu} | \alpha \epsilon_{\lambda_2} e^{-ikr_2} | \psi_i \rangle}{E_{\nu} - E_i + \hbar\omega_2}$$

$$\langle \mathbf{k}_1 \lambda_1, \mathbf{k}_2 \lambda_2 | \hat{\rho}_{2\gamma} | \mathbf{k}_1 \lambda'_1, \mathbf{k}_2 \lambda'_2 \rangle$$

$$\propto \sum_{M_i M'_i M_f} \langle \alpha_i J_i M_i | \hat{\rho}_i | \alpha_i J_i M'_i \rangle \tilde{M}_{fi} \tilde{M}_{fi}^*$$

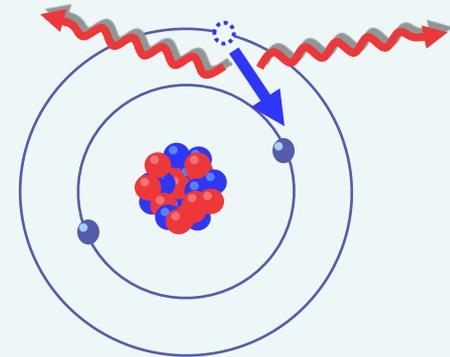
- ▶ Total decay rates
- ▶ Energy (spectral) distributions
- ▶ Angular and polarization correlations
- ▶ Entanglement measures



Two-photon decay of Helium-like ions

- ▶ We have used independent particle model together with the density matrix approach to analyze properties of the two-photon emission from helium-like heavy ions:
 - ⊕ Total and energy-differential decay rates ←
 - ⊕ Angular correlations between emitted photons
 - ⊕ Polarization properties of the photons

Emission angles and polarization states of the photons remain unobserved



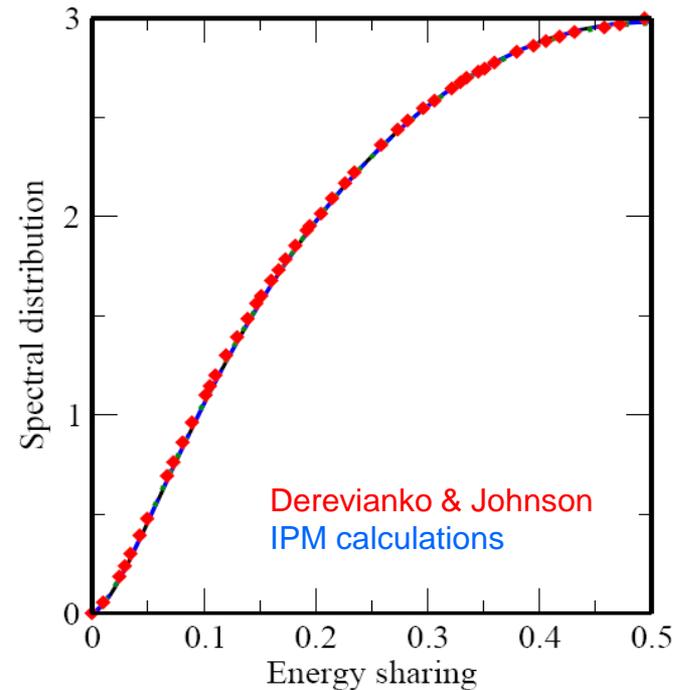
IPM versus Relativistic CI calculations

TABLE II. Two-photon decay rates $w_{2\gamma}$ for 2^1S_0 states of heliumlike ions. Numbers in brackets are powers of ten.

| Z | $w_{2\gamma}$ (s^{-1}) | Z | $w_{2\gamma}$ (s^{-1}) |
|-----|----------------------------|-----|----------------------------|
| 2 | 5.102[01] | 30 | 9.938[09] |
| 3 | 1.940[03] | 35 | 2.540[10] |
| 4 | 1.816[04] | 36 | 3.012[10] |
| 5 | 9.211[04] | 40 | 5.692[10] |
| 6 | 3.300[05] | 41 | 6.604[10] |
| 7 | 9.444[05] | 45 | 1.154[11] |
| 8 | 2.310[06] | 50 | 2.164[11] |
| 9 | 5.029[06] | 54 | 3.415[11] |
| 10 | 1.001[07] | 55 | 3.806[11] |
| 11 | 1.856[07] | 60 | 6.350[11] |
| 12 | 3.249[07] | 65 | 1.013[12] |
| 13 | 5.421[07] | 70 | 1.556[12] |
| 14 | 8.685[07] | 75 | 2.312[12] |
| 15 | 1.344[08] | 80 | 3.336[12] |
| 16 | 2.020[08] | 82 | 3.834[12] |
| 17 | 2.957[08] | 85 | 4.690[12] |
| 18 | 4.230[08] | 90 | 6.439[12] |
| 19 | 5.930[08] | 92 | 7.265[12] |
| 20 | 8.163[08] | 95 | 8.653[12] |
| 25 | 3.249[09] | 100 | 1.140[13] |
| 28 | 6.517[09] | | |

- ▶ Example: Test calculations for the $2S_0 \rightarrow 1S_0$ transition in helium-like uranium.

$$\Gamma_{IPM} = 7.264 \cdot 10^{12} s^{-1}$$



$$x = \frac{\hbar\omega_1}{\hbar\omega_1 + \hbar\omega_2}$$

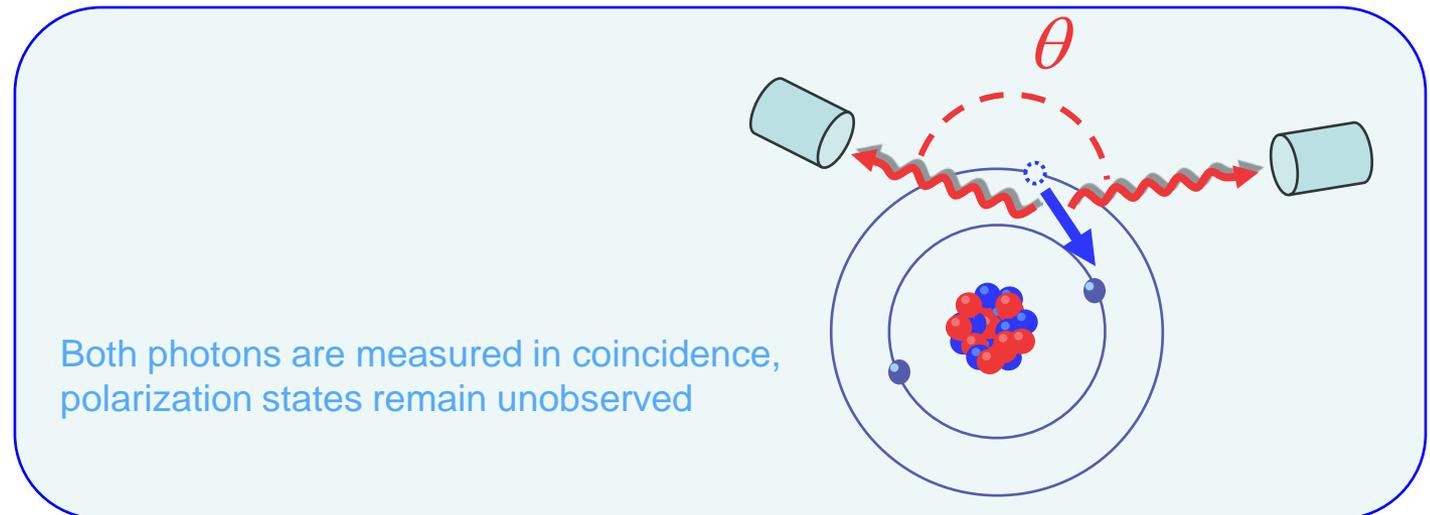
energy sharing
(reduced energy)

A. Derevianko and W. R. Johnson, PRA **58** (1997) 1288

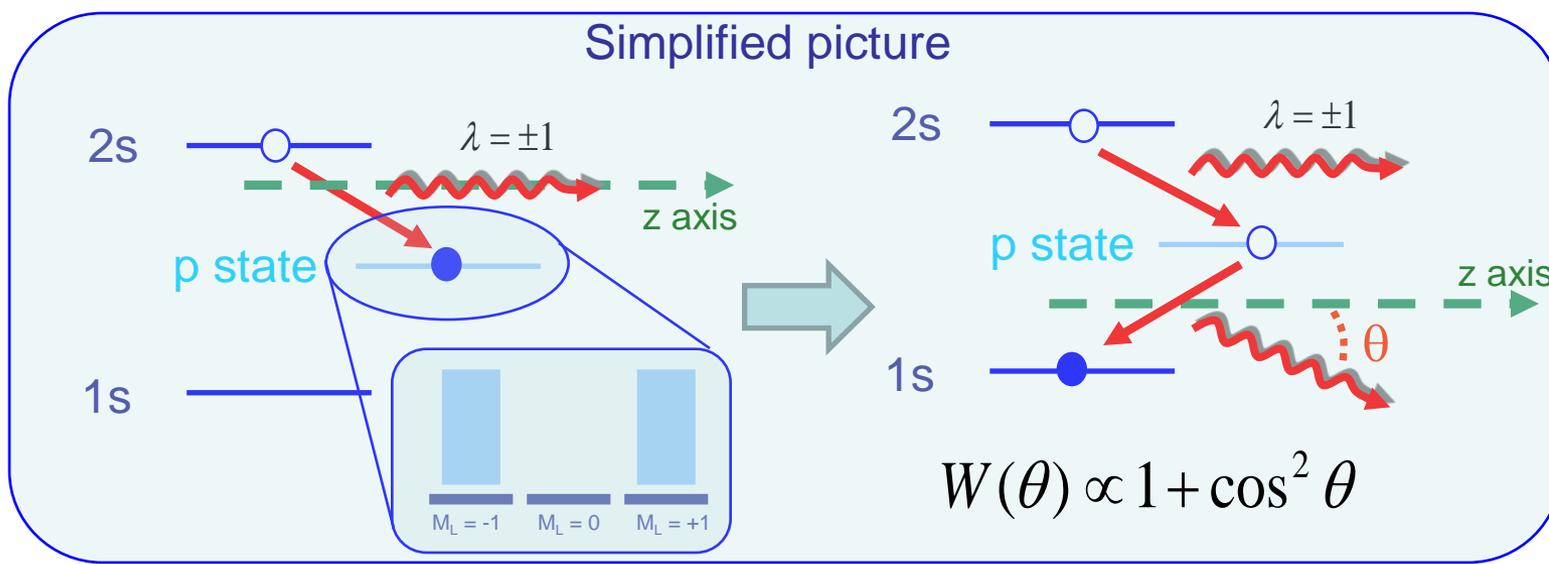
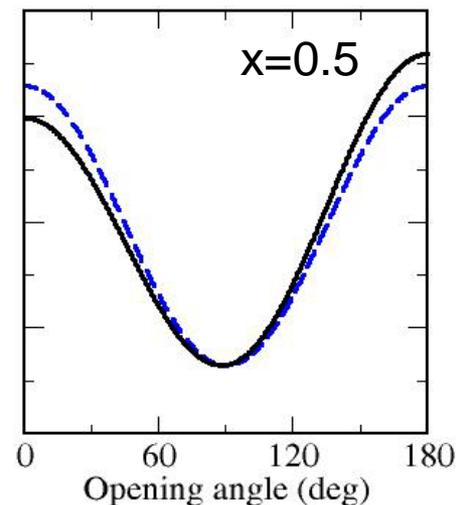
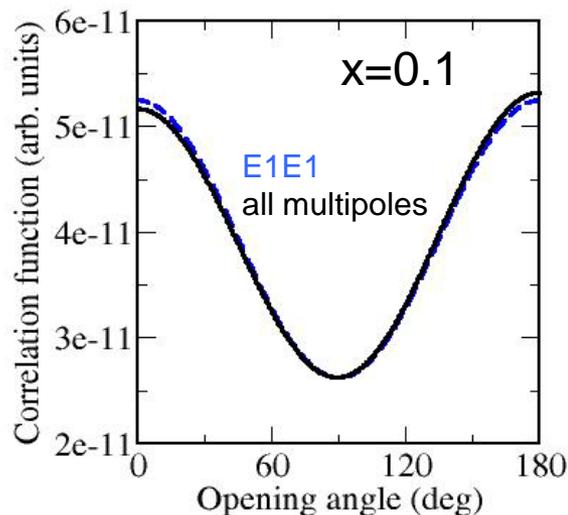
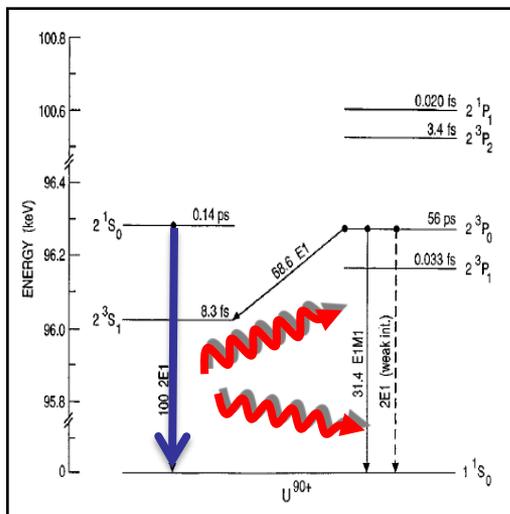
- ▶ Good agreement was found between the IPM and CI calculations!

Two-photon decay of Helium-like ions

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 - ⊕ Total and energy-differential decay rates
 - ⊕ Angular correlations between emitted photons ←
 - ⊕ Polarization properties of the photons

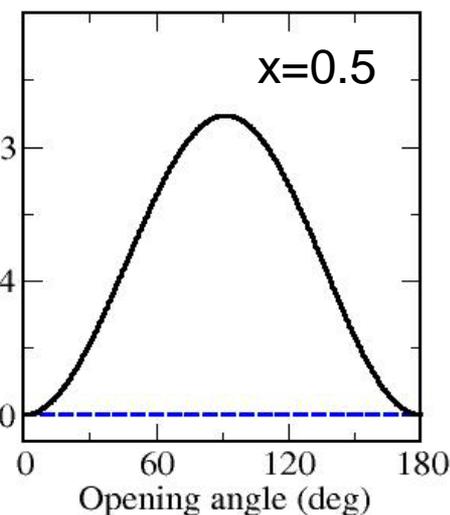
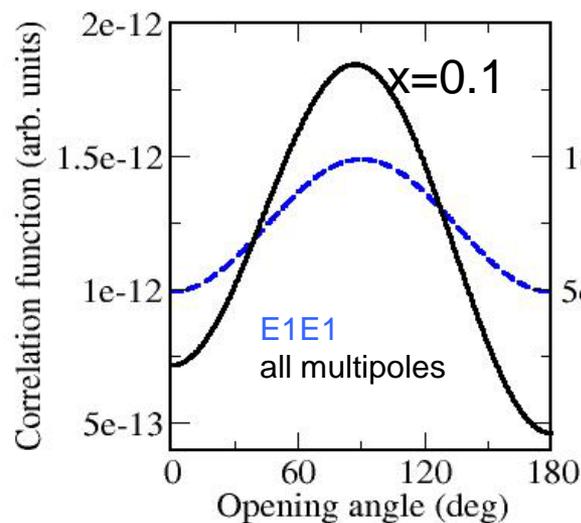
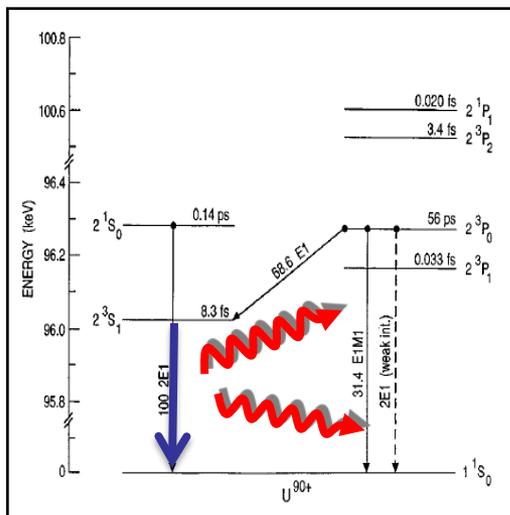


$2S_0 \rightarrow 1S_0$ two-photon decay: Angular correlations



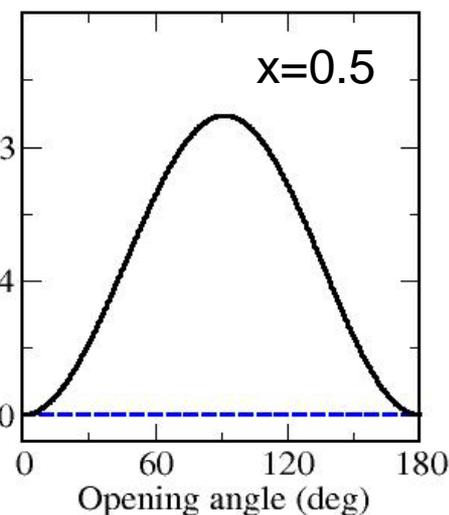
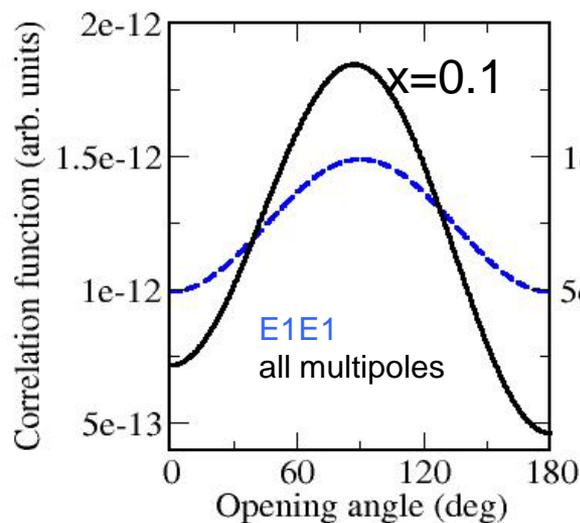
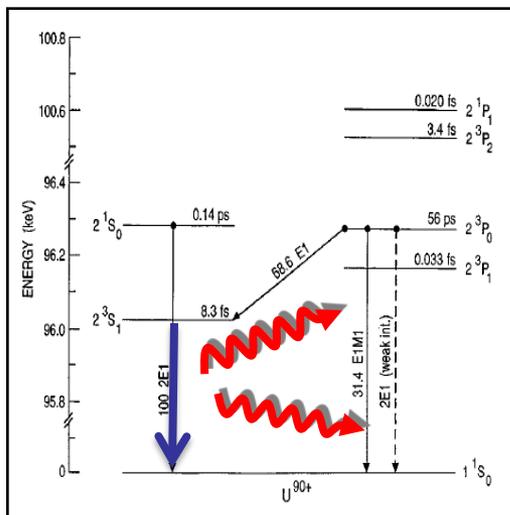
- Non-dipole effects significantly affect angular correlation function!

$2S_1 \rightarrow 1S_0$ two-photon decay: Angular correlations

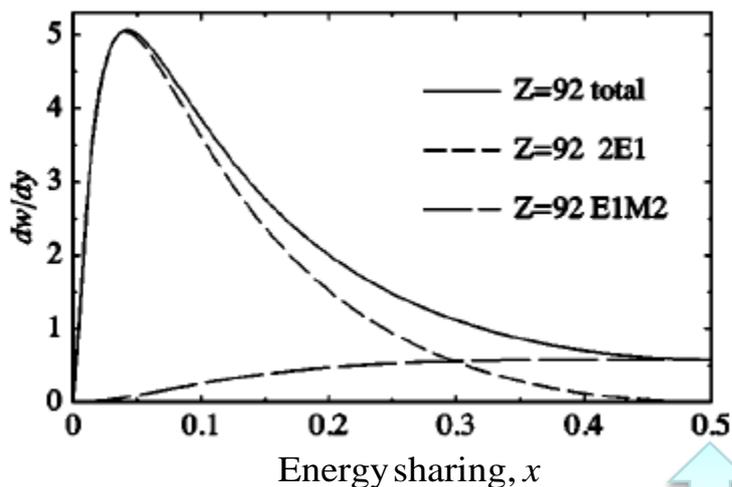


- ▶ One may observe significant higher-multipole effects for $x=0.1$.
- ▶ What happens for the case of equal energy sharing ($x=0.5$)?

$2S_1 \rightarrow 1S_0$ two-photon decay: Angular correlations



- ▶ Similar effect is known for the spectral distribution of emitted photons.

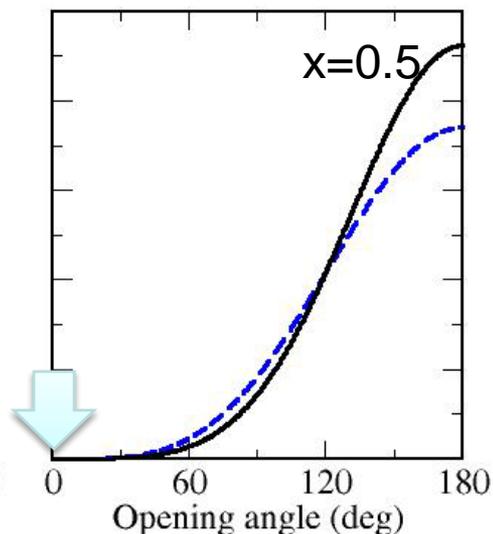
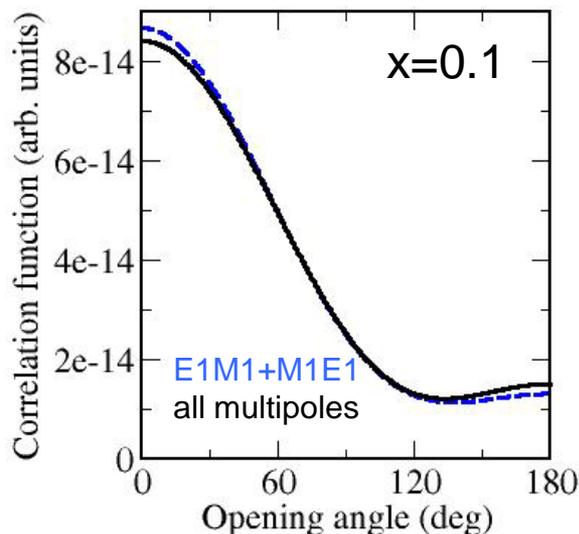
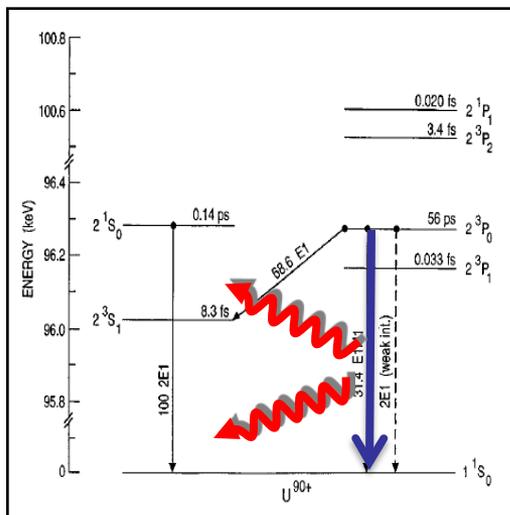


- ▶ It is a result of fundamental symmetry arguments (photons are Bose particles)!
- ▶ Effect is known in particle physics as a Landau-Yang theorem!

Can one use two-photon decay for probe of possible small violations of the spin-statistics relation for photons?



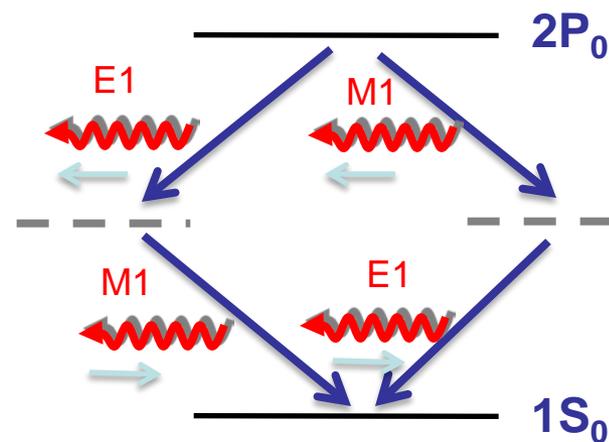
2P₀ → 1S₀ two-photon decay: Angular correlations



- ▶ One may observe significant higher-multipole effects especially for the equal energy sharing.
- ▶ For $x=0.5$ no parallel ($\theta = 0$) emission is possible!

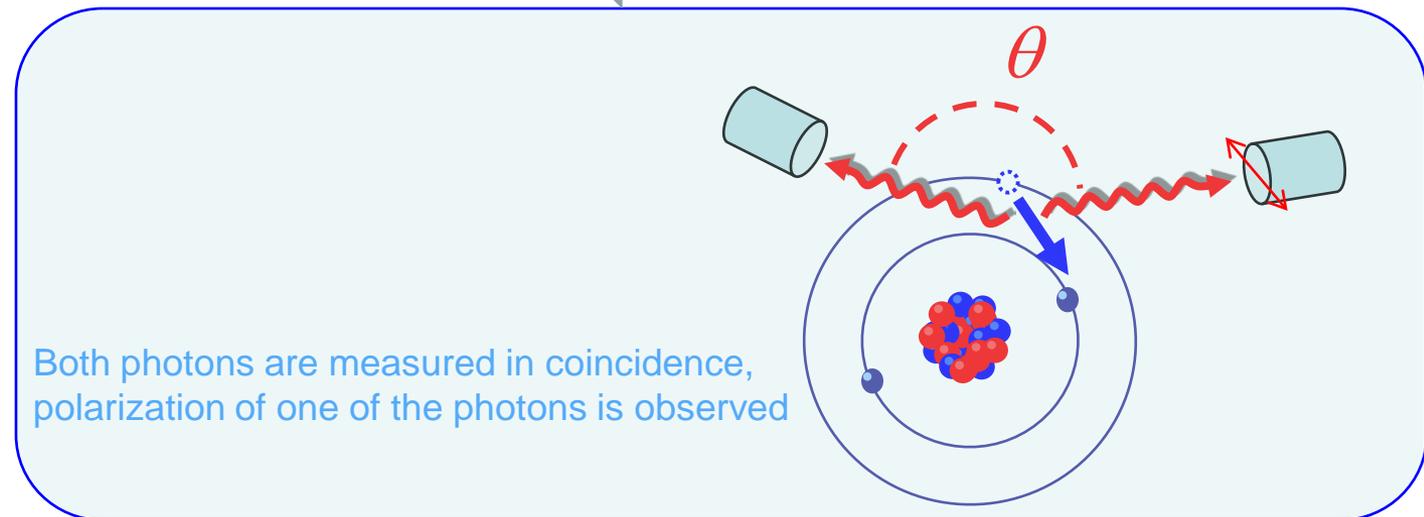
$$\tilde{M}_{fi} \propto \sum_{\nu} \frac{\langle |E1(\lambda)| \rangle \langle |M1(-\lambda)| \rangle}{E_{\nu} - E_i + \hbar\omega} + \frac{\langle |M1(\lambda)| \rangle \langle |E1(-\lambda)| \rangle}{E_{\nu} - E_i + \hbar\omega} = 0$$

- ◆ Interference between two decay channels (double slit-like scheme) suppresses forward emission!

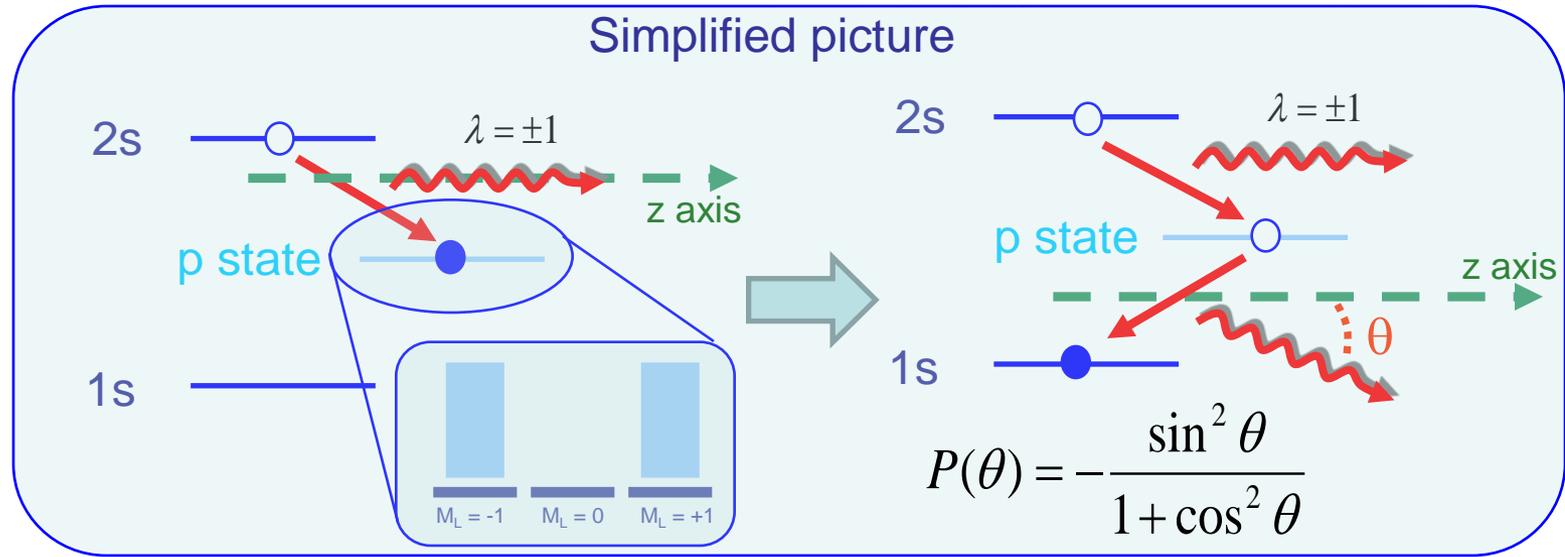
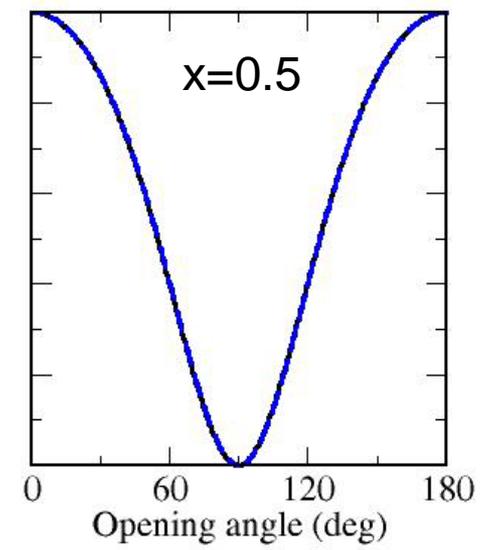
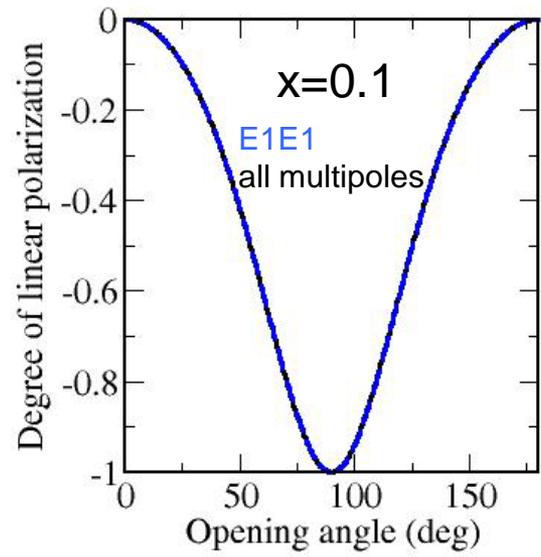
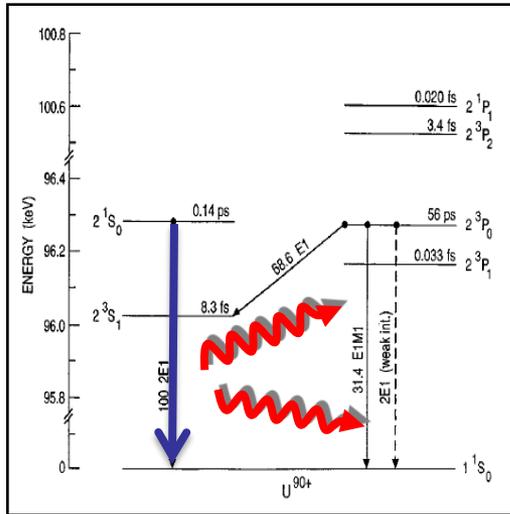


Two-photon decay of Helium-like ions

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 - ⊕ Total and energy-differential decay rates
 - ⊕ Angular correlations between emitted photons
 - ⊕ Polarization properties of the photons ←



$2S_0 \rightarrow 1S_0$ two-photon decay: Linear polarization



► Non-dipole effects seem to be negligible for linear polarization. But why?

$2S_0 \rightarrow 1S_0$ two-photon decay: Linear polarization

In the non-relativistic dipole approximation two emitted photons are in *pure quantum state*:

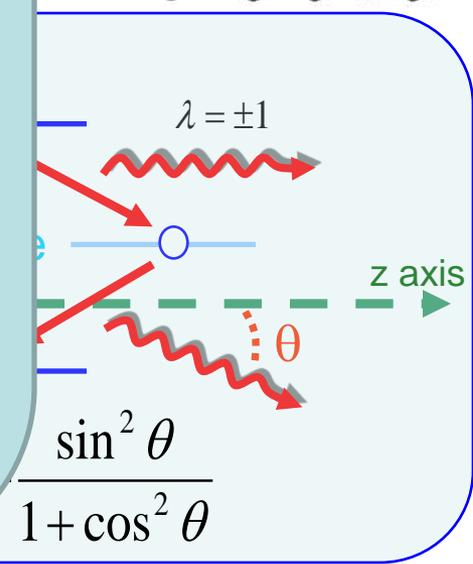
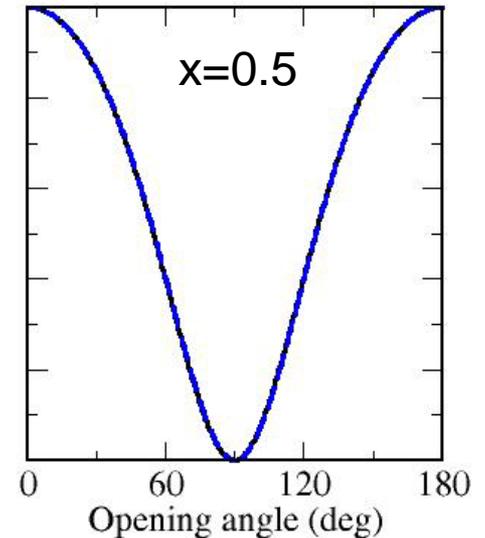
$$|\Psi_{12}\rangle = \frac{1}{\sqrt{1 + \cos^2 \theta}} (|y_1 y_2\rangle + \cos \theta |x_1 x_2\rangle)$$

- For $\theta = 0$ and 180 this state is fully entangled (Bell state) and, hence, after tracing over the quantum numbers of one of the photons we find the other one to be in *fully mixed state*:

$$\hat{\rho}_1 = (|x_1\rangle\langle x_1| + |y_1\rangle\langle y_1|) / 2$$

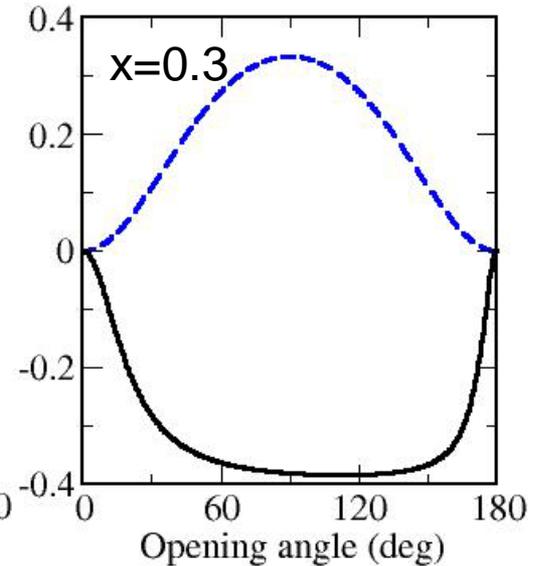
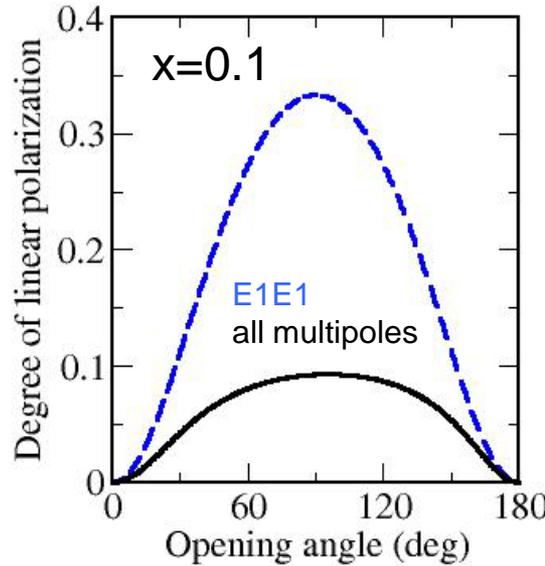
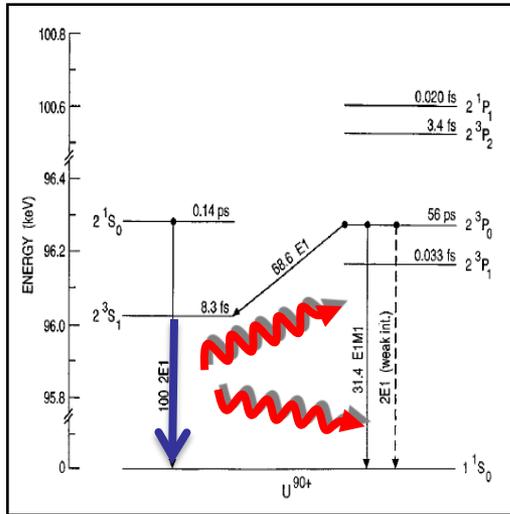
- In contrast, for $\theta = 90$ photons are in product (unentangled) state. After taking trace over unobserved quantum numbers “our” photon is still in the *pure state*:

$$\hat{\rho}_1 = |y_1\rangle\langle y_1|$$

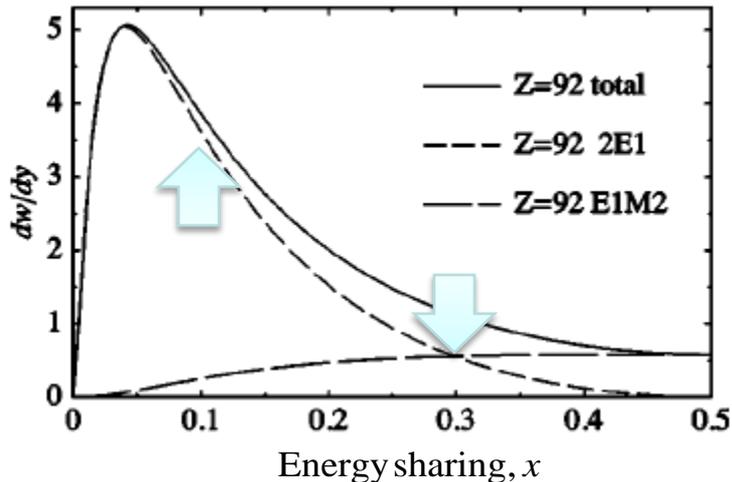


- Non-dipole effects seem to be negligible for linear polarization. But why?

$2S_1 \rightarrow 1S_0$ two-photon decay: Linear polarization



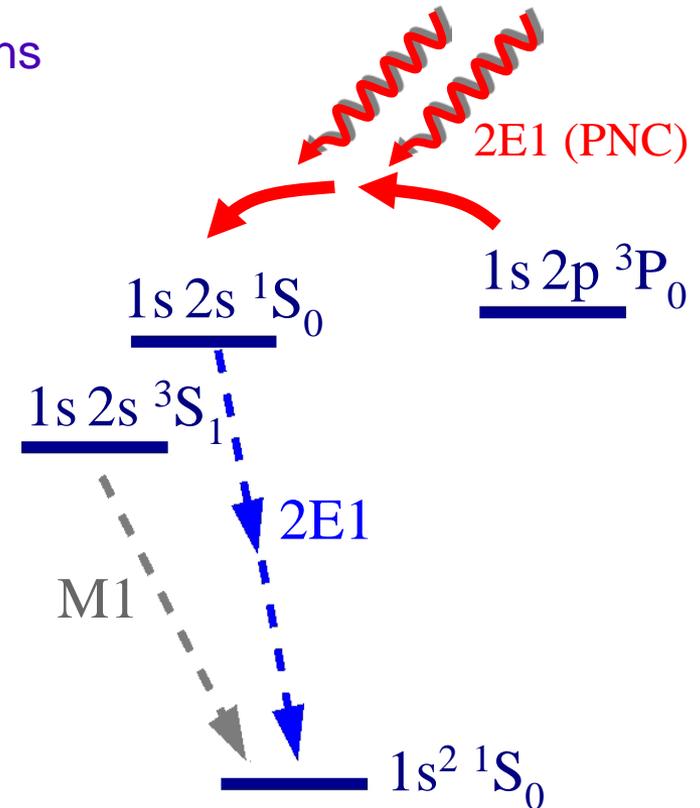
Preliminary results!



- ▶ Polarization properties of the two-photon decay of the triplet $2S$ state are strongly affected by the non-dipole effects!
- ▶ For high- Z ions, this decay channel is not dominant one!

Summary and outlook

- ▶ We have used independent particle model together with the density matrix approach to analyze properties of the two-photon emission from helium-like ions:
 - ⊕ Total and energy-differential decay rates
 - ⊕ Angular correlations between emitted photons
 - ⊕ Polarization properties of the photons
- ▶ Our next step is analysis of parity-violating two-photon transitions in helium-like Uranium!



Thank you for your attention!