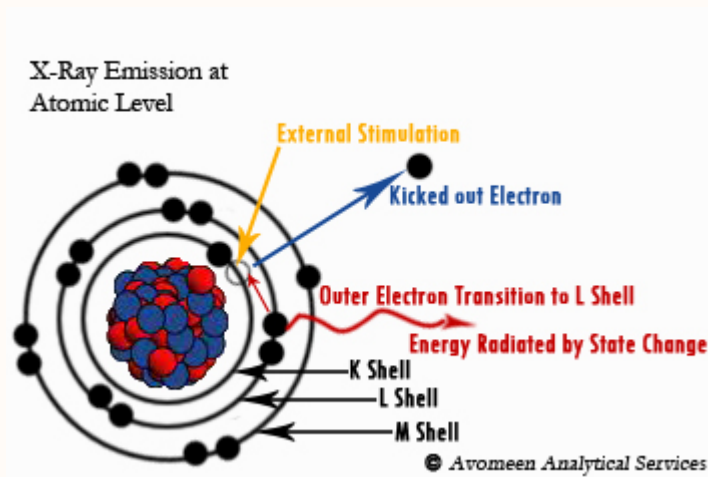


# High energy PIXE: measurements for Cu, Ag and Ti K-shell ionization cross sections and comparisons with theoretical model

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# Particle Induced X-ray Emission (PIXE)



- PIXE method :
  - Incident beam energy :  $<4$  MeV
  - Non-destructive multi-elemental method ( $Z > 11$ )
  - Fast analysis (few minutes)
  - Quantitative analysis
  - Good sensitivity ( $\mu\text{g/g}$ )
  - Analysis of thin layers close to the surface

# High energy PIXE (HE PIXE)

- study of thick and multilayer objects:
  - Energy beam: few MeV
  - Proton range:  $\sim$ mm
  - In-depth analysis
  - Adapted for thick archeologic object
  - Detection of elements thanks to their more energetic K line



A. Denker et al, Nucl. Inst. Meth. B 239 (2005) 65–70

# Quantitative analysis (PIXE) for the element (Z) in a matrix

Number of detected X-rays      Number of atoms      Intensity of X-line      Ionization cross section      Transmission factor

$$N_X^Z = N_p N^Z \omega b \varepsilon \int_{E_f}^{E_0} \frac{\sigma_i(E) \times T(E)}{S(E)} dE$$

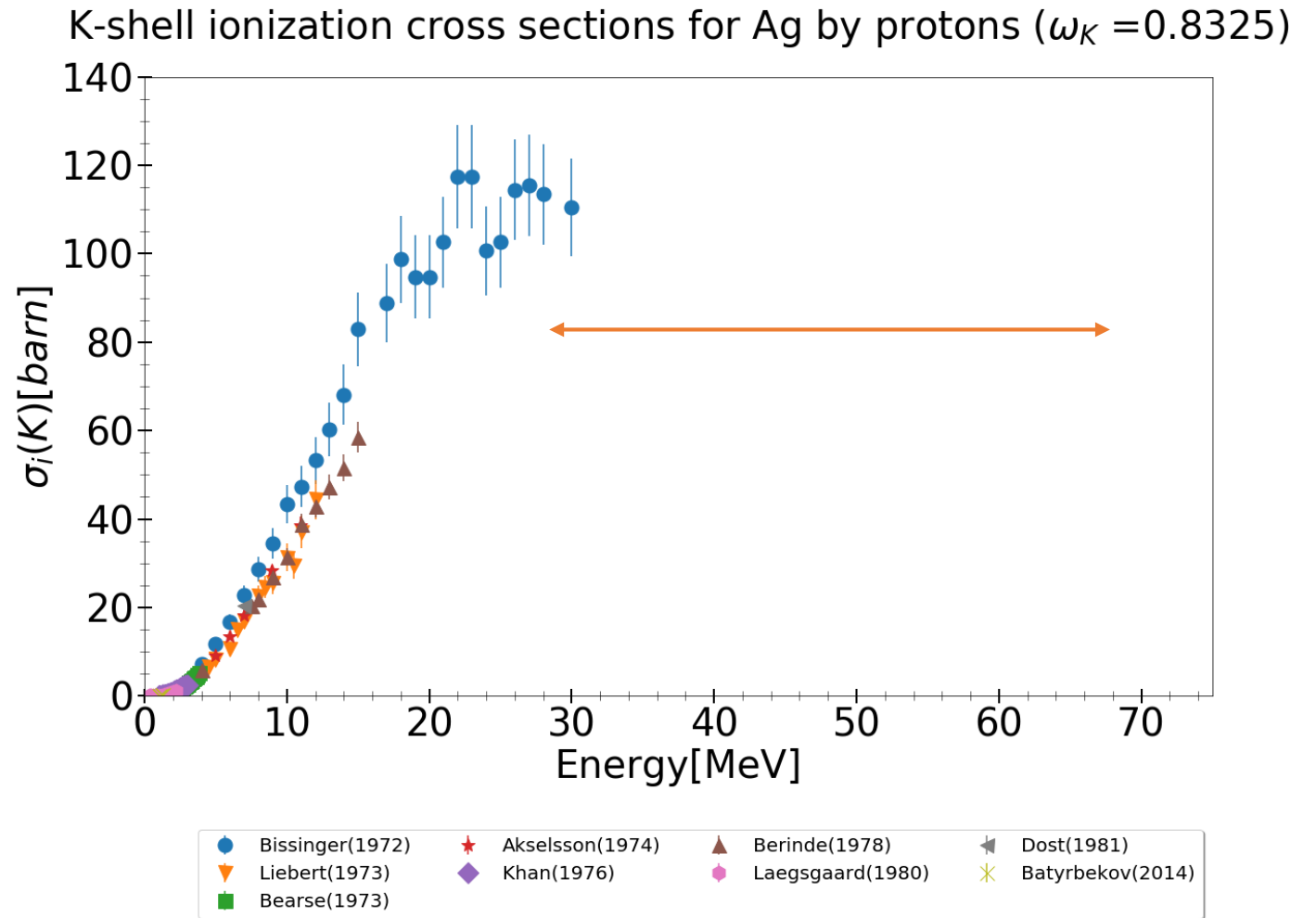
Number of incident particles      Fluorescence yields      Detection efficiency (intrinsic + geometric)      Stopping power

Quantization requires a good knowledge of the ionization cross section :

- Experimental data
- Theoretical values

# Ionization cross section: Experimental data available

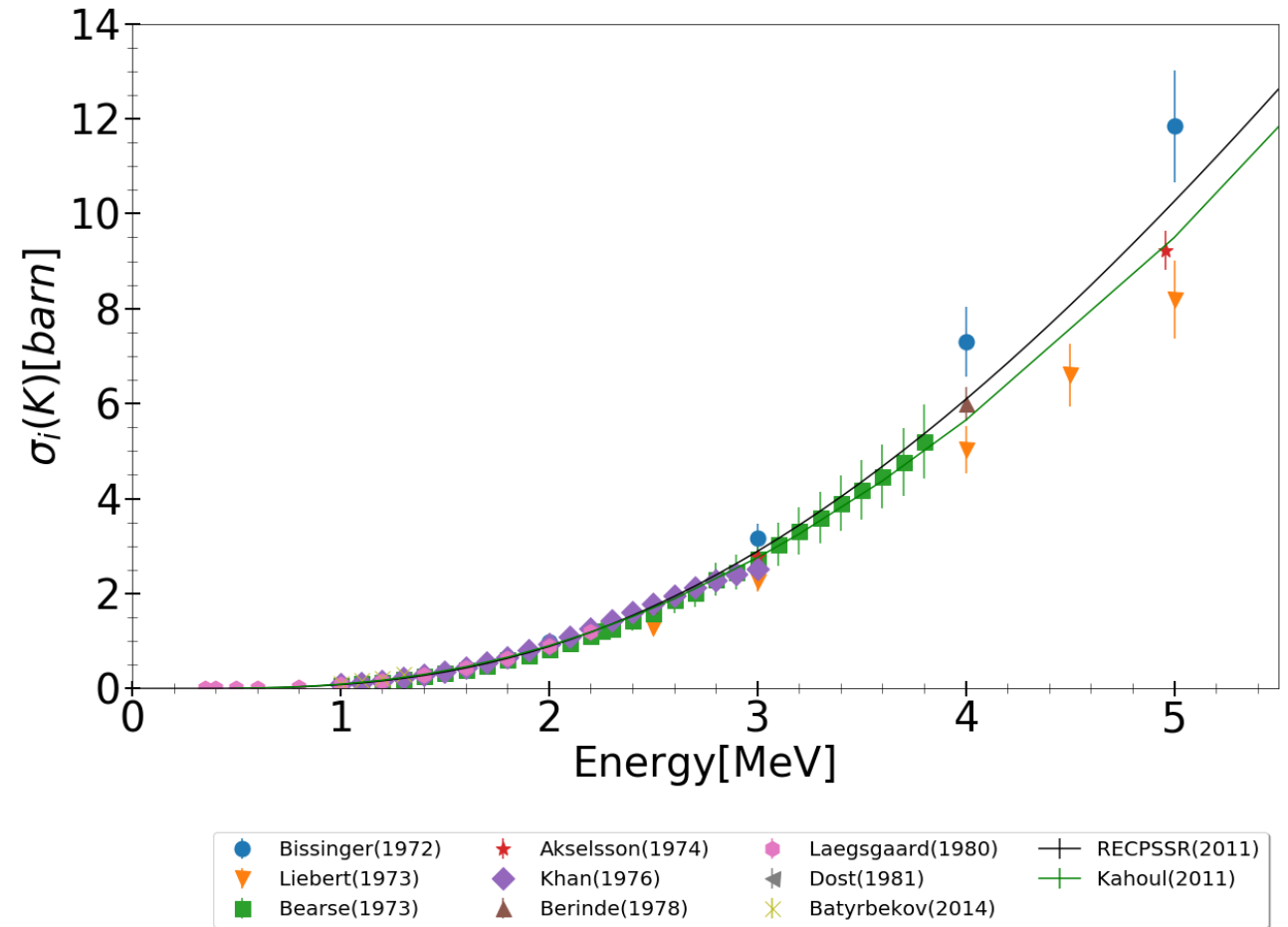
- Experimental data at high energy are scarce



# Ionization cross section: Theoretical model (RECPSSR)

- RECPSSR model takes into account the relativistic effect of the projectile

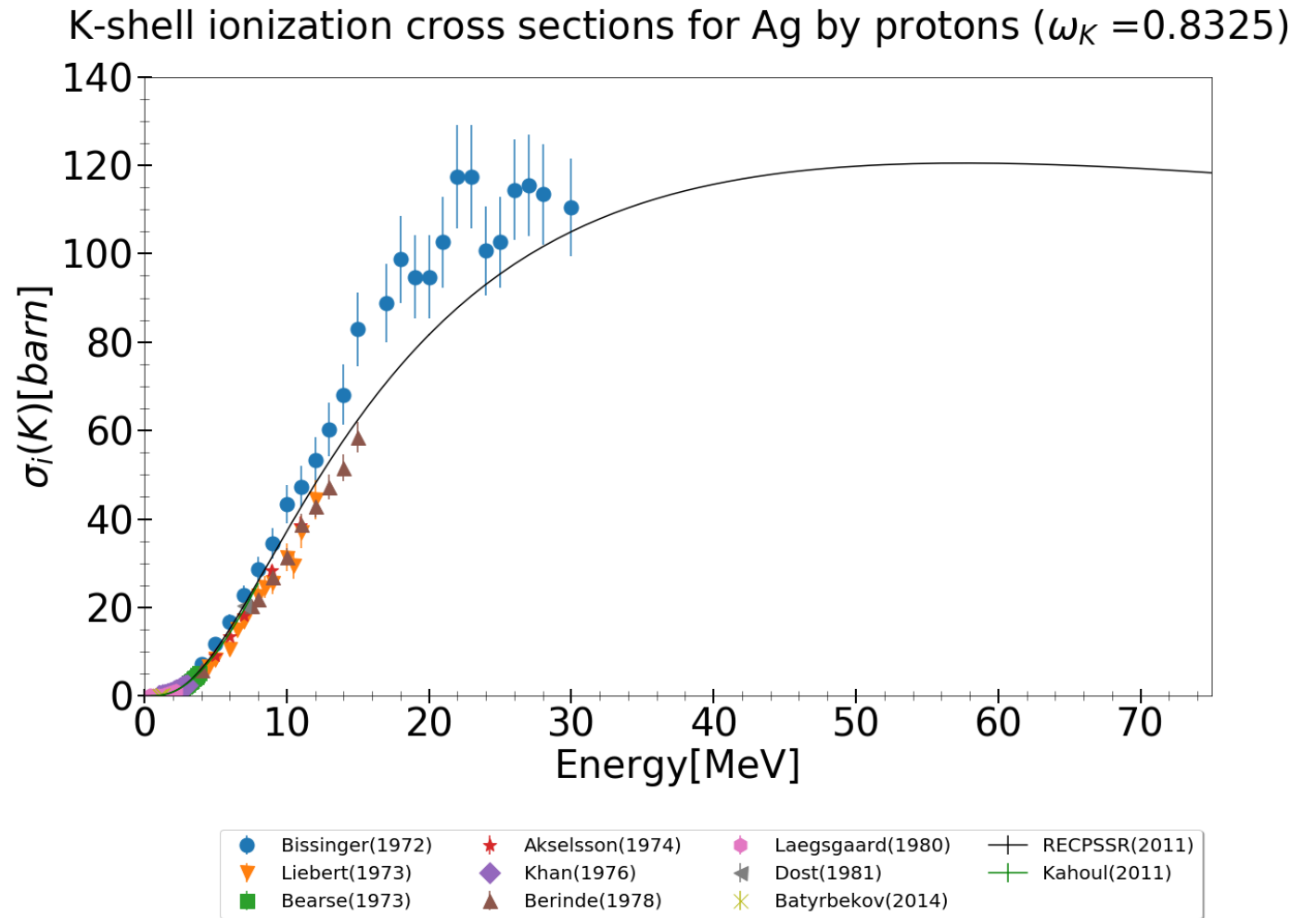
K-shell ionization cross sections for Ag by protons ( $\omega_K = 0.8325$ )



A. Kahoul et al., Rad. Phy. And Chem. 80 (2011) 1300-1311  
W. Brandt et al., Phys. Rev. A 23 (1981) 1717

# Ionization cross section: Theoretical model (RECPSSR)

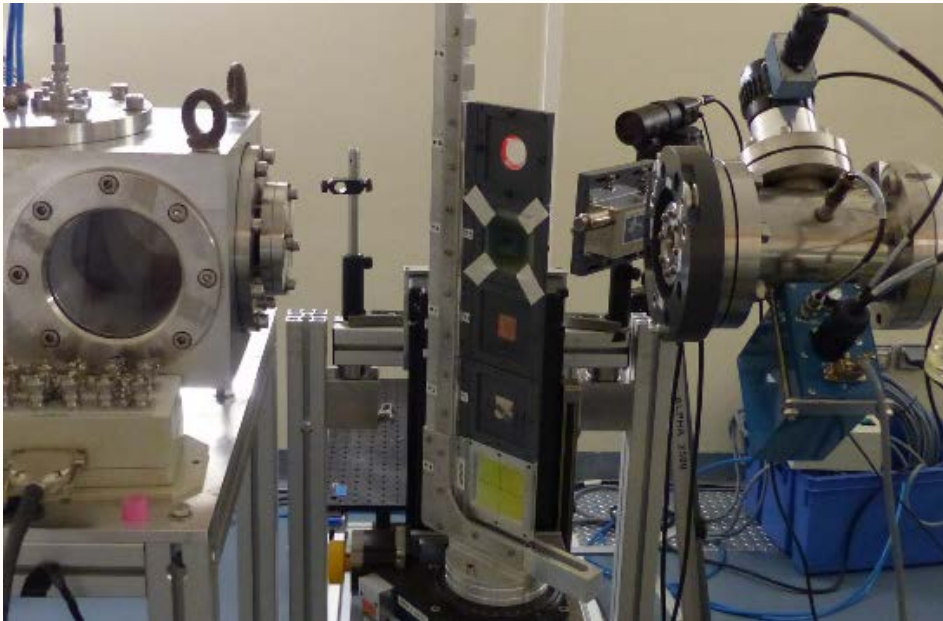
- RECPSSR model takes into account the relativistic effect of the projectile
- RECPSSR model have been validated with the bibliographic data at low energy
- The lack of high-energy experimental data does not allow to verify the model over this range in energy (30 – 70 MeV)



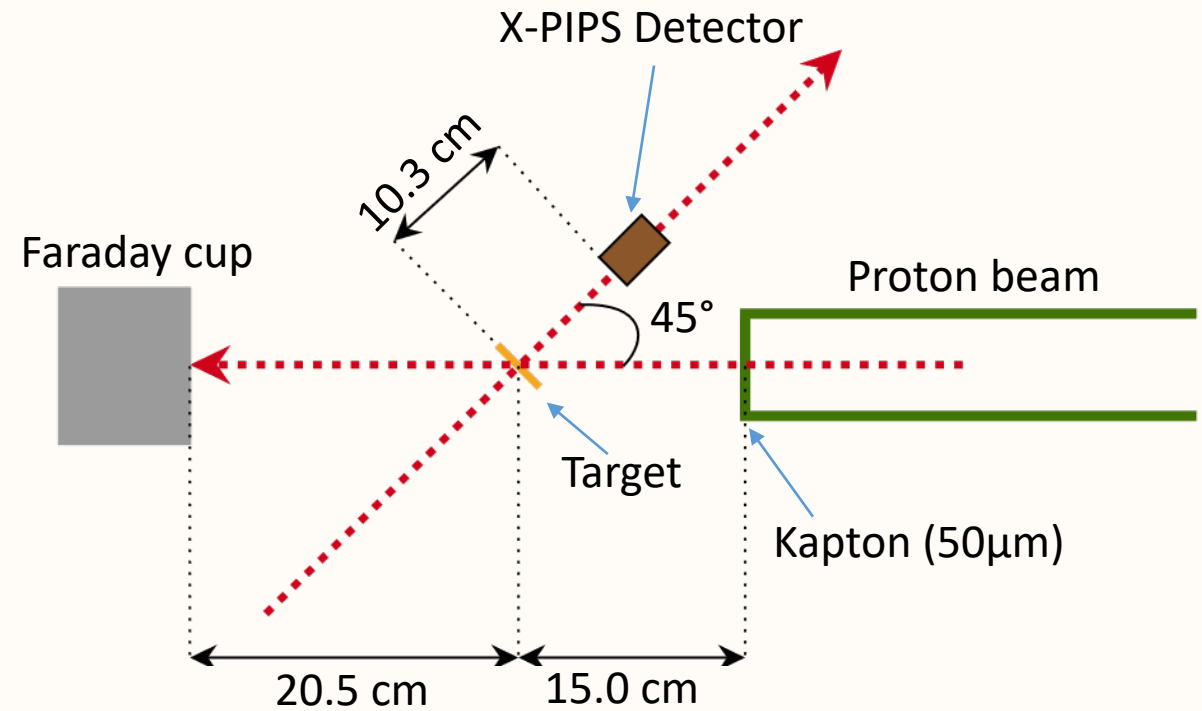
A. Kahoul et al., Rad. Phy. And Chem. 80 (2011) 1300-1311  
W. Brandt et al., Phys. Rev. A 23 (1981) 1717



# Experimental measurements for X-ray production cross section:



Cyclotron C70 ARRONAX, Nantes, France



Thin targets (10  $\mu$ m):  
Titanium (Ti), Copper (Cu) and Silver (Ag)

Particles: Protons  
Energies: 35, 40, 50 and 68 MeV



$$N_X^Z = N_p N^Z \omega b \varepsilon \int_{E_f}^{E_0} \frac{\sigma_i(E) \times T(E)}{S(E)} dE$$

# Experimental set-up

## K X-rays production cross section

- Thin target → constant production cross section
- Production cross section → Independent of the fluorescence yield
  - For each K-lines  $\sigma_P^T(K) = \sum_i \sigma_P(K_i)$  → Independent of the intensity of a given transition b

$$\sigma_P(K_i) = \frac{N_X(K_i) \times M \times \frac{\mu}{\rho}(K_i) \times \rho \times \cos(45)}{N_P \times \varepsilon_{tot}(K_i) \times N_A \times \rho \times \left(1 - \exp\left(-\frac{\mu}{\rho}(K_i) \times \rho \times d\right)\right)}$$

Number of X-rays detected (X-PIPS detector) →  $N_X(K_i)$   
 Atomic mass →  $M$   
 Mass attenuation coefficient →  $\frac{\mu}{\rho}(K_i)$   
 $\cos(45)$   
 K<sub>i</sub> X-rays production cross sections →  $\sigma_P(K_i)$   
 Number of incident particles (Faraday cup) →  $N_P$   
 Detection efficiency →  $\varepsilon_{tot}(K_i)$   
 Avogadro number →  $N_A$   
 $\rho$   
 $\left(1 - \exp\left(-\frac{\mu}{\rho}(K_i) \times \rho \times d\right)\right)$

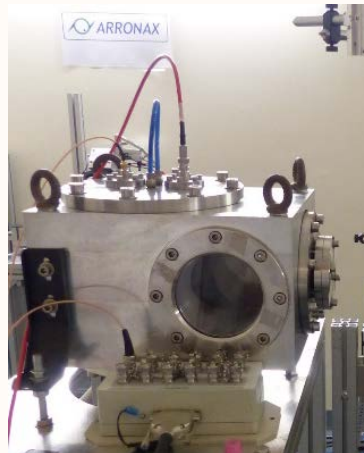
# Experimental set-up

$$\sigma_p(K_i) = \frac{N_X(K_i) \times M \times \frac{\mu}{\rho}(K_i) \times \rho \times \cos(45)}{N_p \times \varepsilon_{tot}(K_i) \times N_A \times \rho \times \left(1 - \exp\left(-\frac{\mu}{\rho}(K_i) \times \rho \times d\right)\right)}$$

## Current load measurement

- Use of an faraday cup equipped with a guard ring
- Uncertainty : 0.5%
- For our measurements, all incident particles reach the faraday cup

→ Number of incident particles  $N_p$



## Thin targets

- Determination of the thickness:
  - Mass with a precision of  $10^{-5}$ g
  - Surface with a high-precision scanner
- Titanium:  $9.95 \pm 0.09 \mu\text{m}$
- Copper:  $10.38 \pm 0.09 \mu\text{m}$
- Silver:  $10.11 \pm 0.09 \mu\text{m}$

→ Thickness of the target  $d$

# Experimental set-up

$$\sigma_p(K_i) = \frac{N_X(K_i) \times M \times \frac{\mu}{\rho}(K_i) \times \rho \times \cos(45)}{N_p \times \epsilon_{tot}(K_i) \times N_A \times \rho \times \left(1 - \exp\left(-\frac{\mu}{\rho}(K_i) \times \rho \times d\right)\right)}$$

## Detection efficiency $\epsilon_{tot}(K_i)$

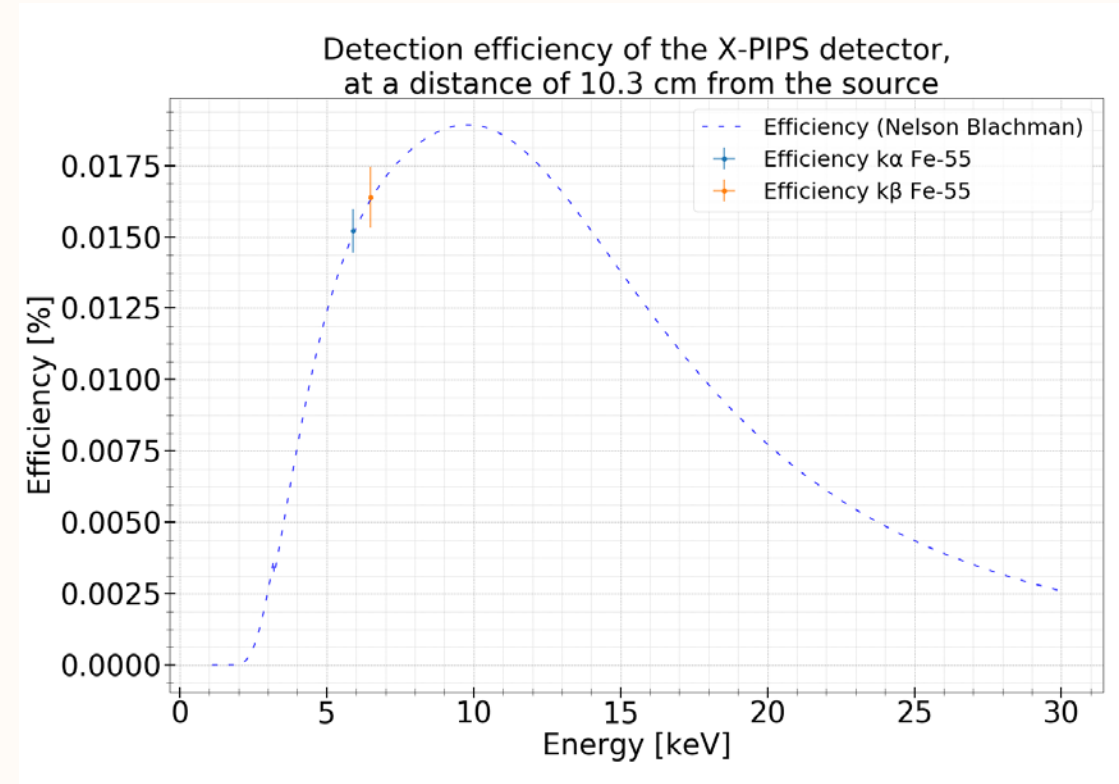
- Use of an Fe-55 source for calibration and validation of theoretical efficiency model (Nelson-Blachmann)

A.H. Jaffrey, Rev. Sci. Instrum 25 (1954) 349  
H.R.V. Carrillo, Nucl. Instr. And Meth. A 371 (1996) 353

- Uncertainty : 5%
- X-PIPS detector : SDD (500 $\mu$ m, 30mm<sup>2</sup>)



Tomography scan of X-PIPS detector

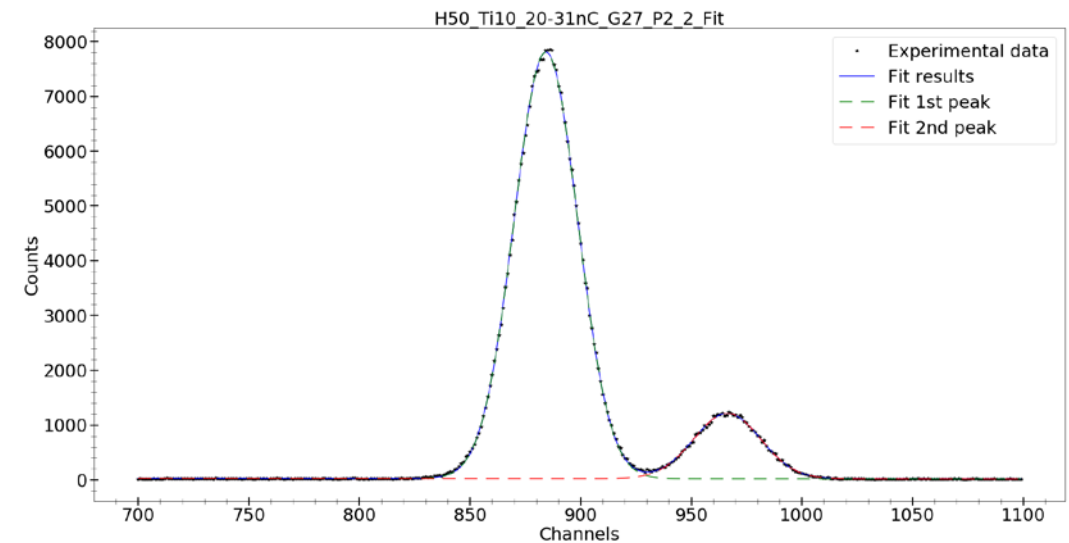


# Experimental set-up

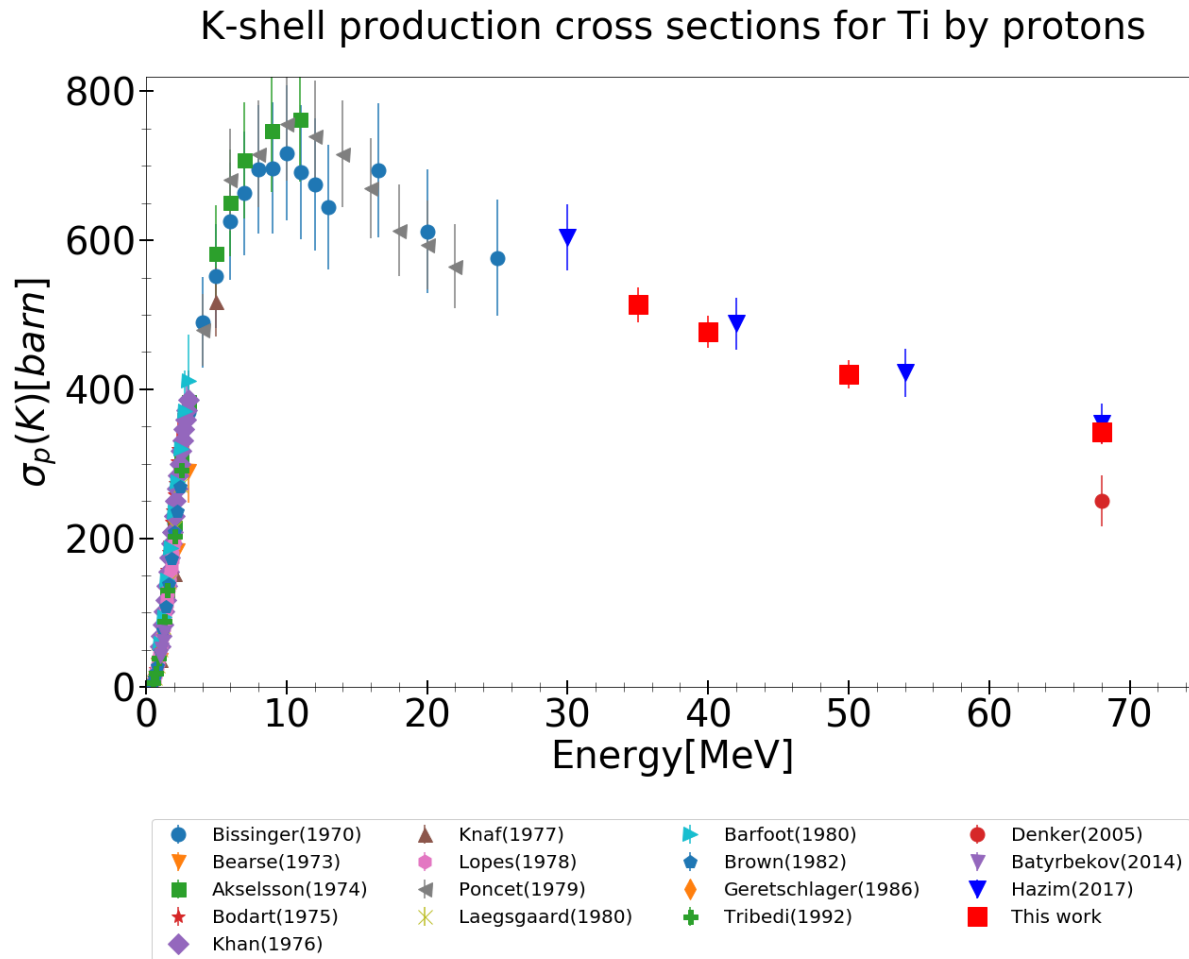
$$\sigma_p(K_i) = \frac{N_X(K_i) \times M \times \frac{\mu}{\rho}(K_i) \times \rho \times \cos(45)}{N_p \times \varepsilon_{tot}(K_i) \times N_A \times \rho \times \left(1 - \exp\left(-\frac{\mu}{\rho}(K_i) \times \rho \times d\right)\right)}$$

## Measurement of the number of X-rays $N_X(K_i)$

- Use of the X-PIPS detector
- Number of X-rays obtained after :
  - fitting of the characteristic peaks of the K layers.
  - integration on fitting curve
  - Correction dead time of detector
- Uncertainty : <1%
  - (statistic and fitting)



# Experimental results: K X-rays production cross sections for Titanium

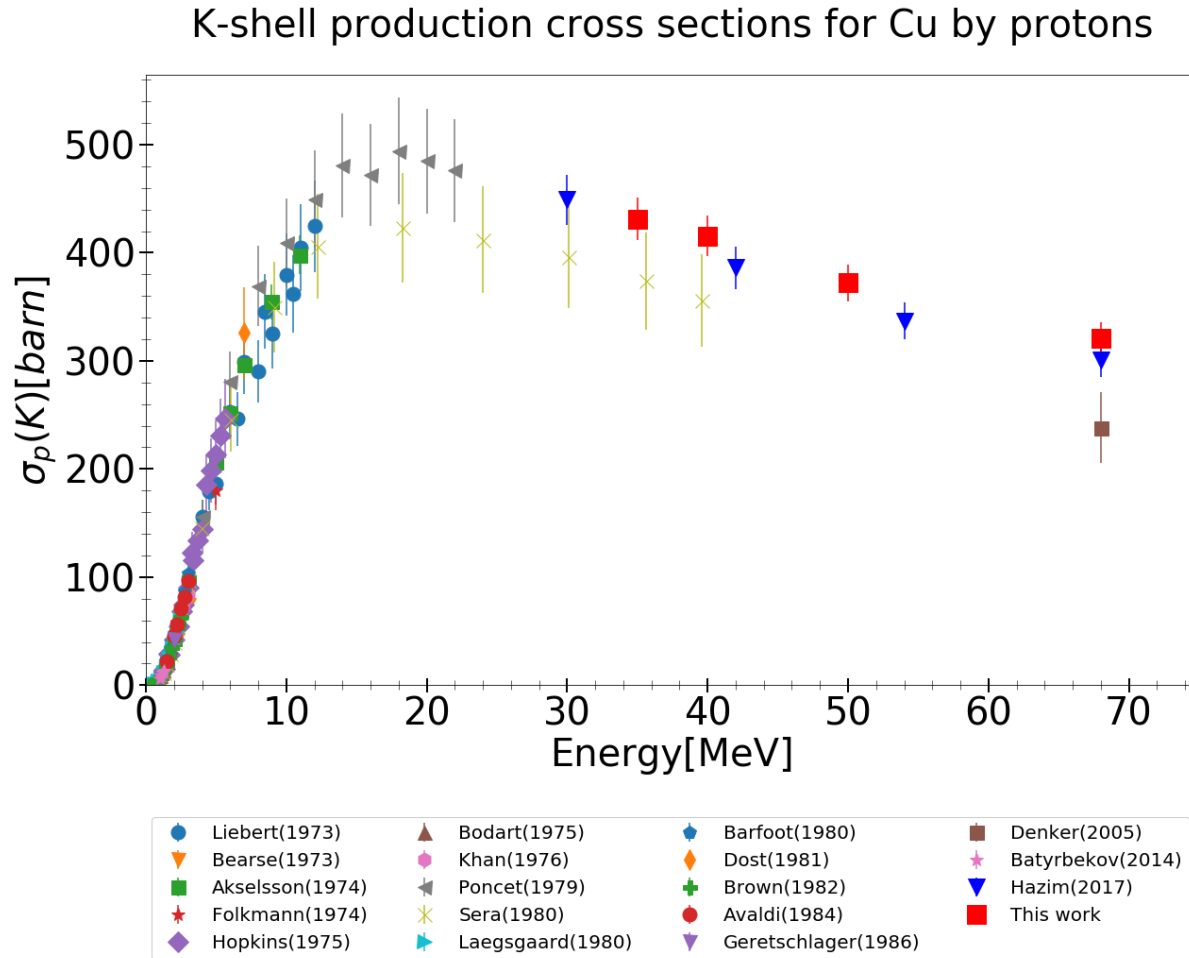


- Our measurements (red square) are
  - consistent with the low-energy trend
  - above the experimental point at 68 MeV
  - Good matching with previous data acquired with a Ge detector (blue triangle)

M. Hazim, PhD thesis, Université de Nantes, march 2017

# Experimental results:

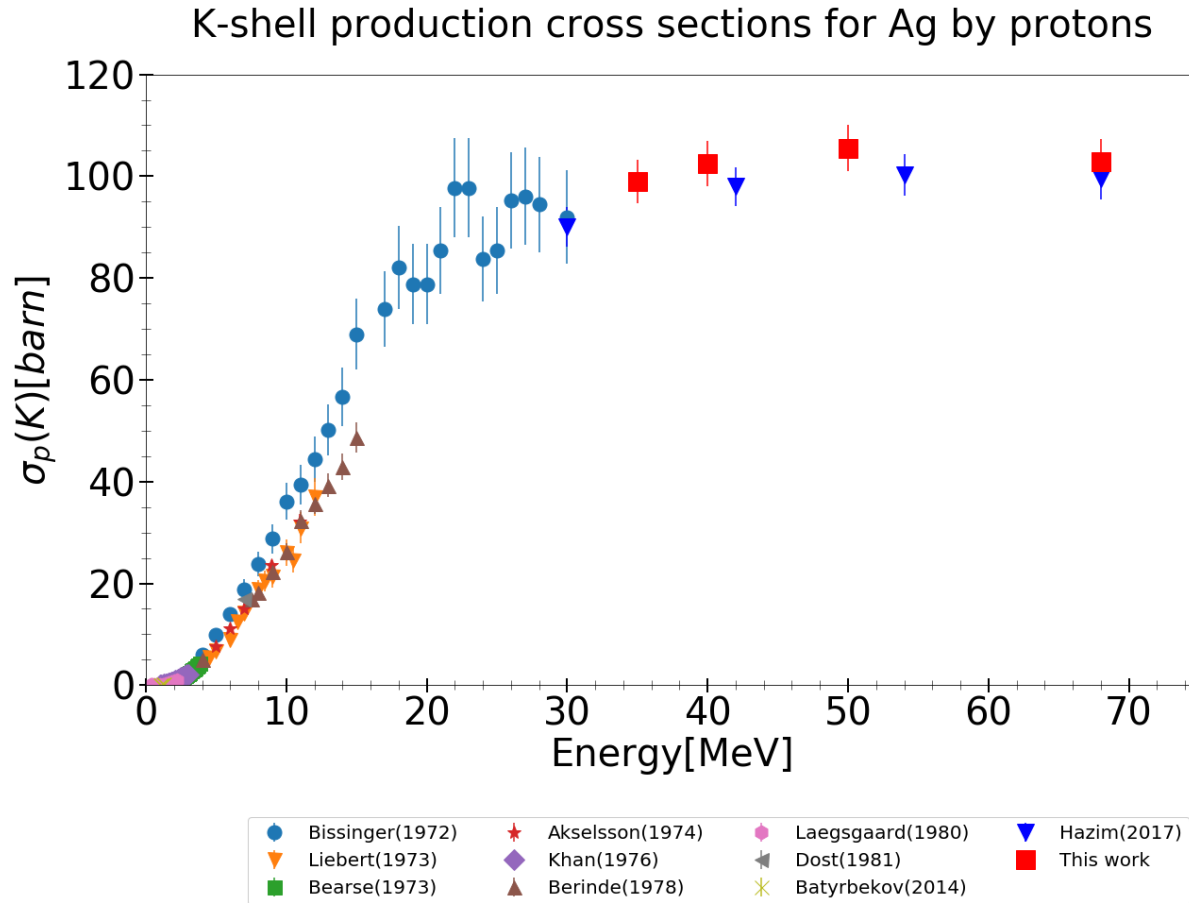
## K X-rays production cross sections for Copper



- Our measurements (red square) are
  - consistent with the low-energy trend
  - above the experimental point at 68 MeV
  - Good matching with previous data acquired with a Ge detector (blue triangle)

# Experimental results:

## K X-rays production cross sections for Silver



- Our measurements (red square) are
  - consistent with the low-energy trend
  - Good matching with previous data acquired with a Ge detector (blue triangle)

What about models ?



# Production/Ionization cross section

K-shell ionization cross section

K-shell production cross section

$$\sigma_I(K) = \frac{\sigma_P^T(K)}{\omega_Z}$$

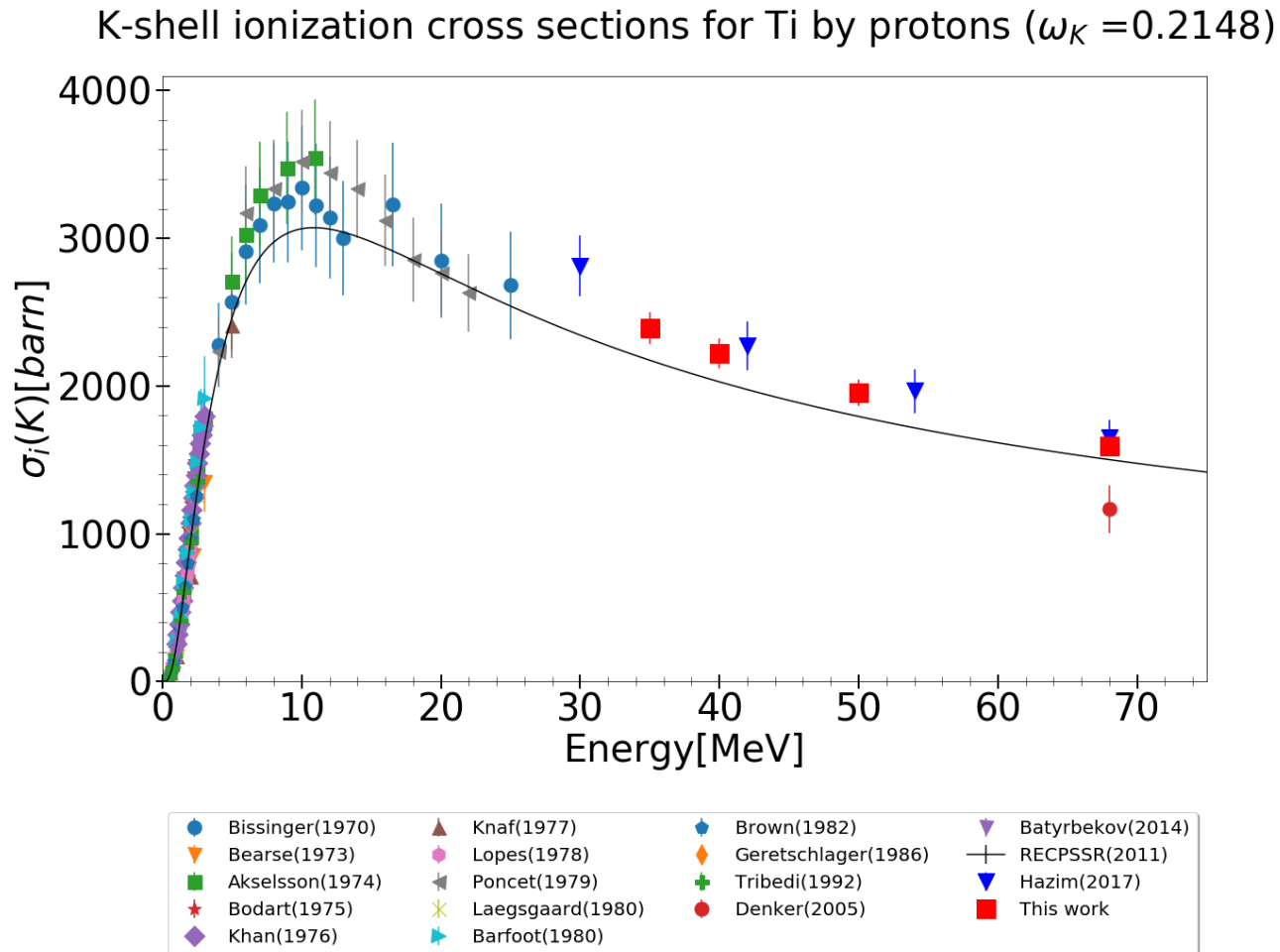
Florescence yield

The fluorescence yield used is an average of the bibliographic data weighted by the errors

$$\omega_K = \frac{1}{\sum_{i=1}^N \left(\frac{1}{\Delta\omega_i}\right)^2} \sum_{i=1}^N \left(\frac{\omega_i}{(\Delta\omega_i)^2}\right)$$

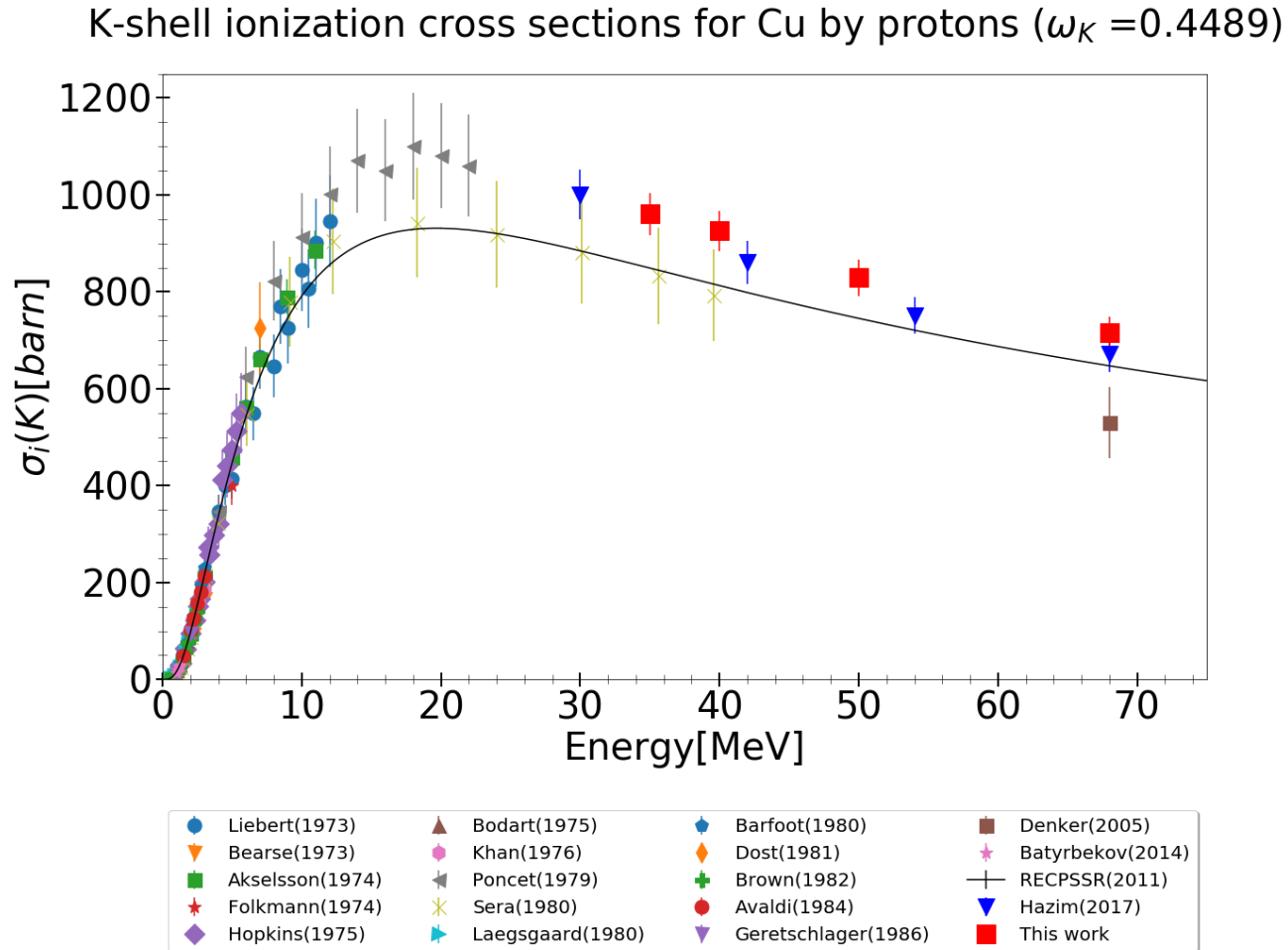
Target	$\omega_K$
Ti	0,2148 ± 0,0050
Cu	0,4489 ± 0,0060
Ag	0,8320 ± 0,0102

# Comparison with the theory (RECPSSR): K X-rays ionization cross sections for Titanium



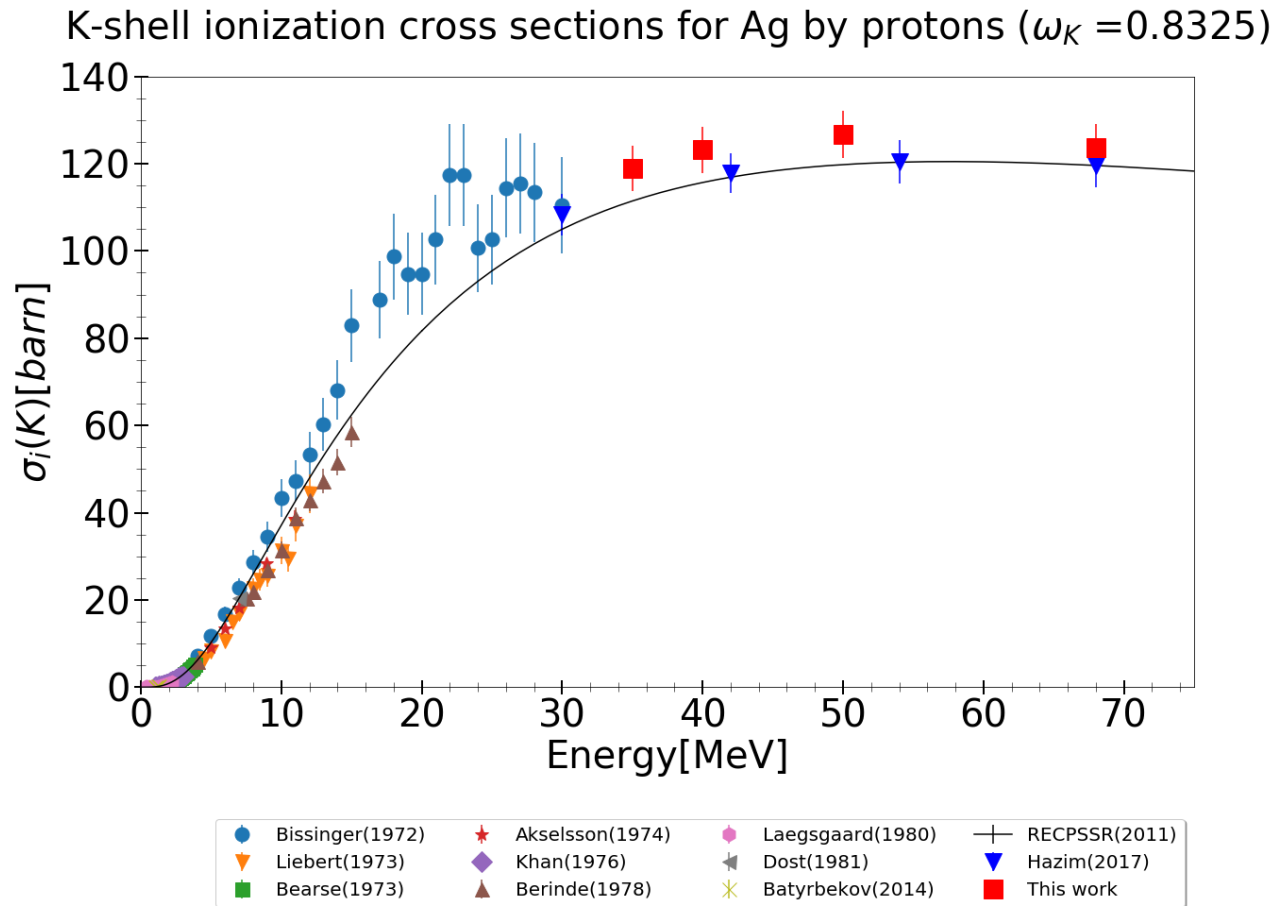
- The model correctly reproduces the trend of the experimental points
- But slightly lower than our experimental data

# Comparison with the theory (RECPSSR): K X-rays ionization cross sections for Copper



- The model correctly reproduces the trend of the experimental points
- But lower than our experimental data

# Comparison with the theory (RECPSSR): K X-rays ionization cross sections for Silver



- The model correctly reproduces the trend of the experimental points
- But slightly lower than our experimental data

# Conclusions:

- We have measured ionization cross section with high-energy protons (30 – 68 MeV)
- It completes the lack of experimental data in this energy region
  
- We have compared our measurements with a theoretical model
  - At high-energy, there is an underestimation of the ionization cross section
  
- what remains to be done:
  - acquire new experimental data with other targets and energies
  - understand the difference between our experimental points and the theoretical model
  - to develop HE PIXE applications at the ARRONAX cyclotron.

# Thanks for your attention

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