Understanding the x-ray emission spectrum after excitation with a source of x-rays: from theory to experiment

Jorge E. Fernández[1,2]

[1]Laboratory of Montecuccolino-DIN, Alma Mater Studiorum University of Bologna, via dei Colli 16, 40136 Bologna, Italy

[2] Alma Mater Studiorum Università di Bologna Representacion en la Republica Argentina, M. T. de Alvear 1149, Buenos Aires, Argentina

Outline

- Description of x-ray emission

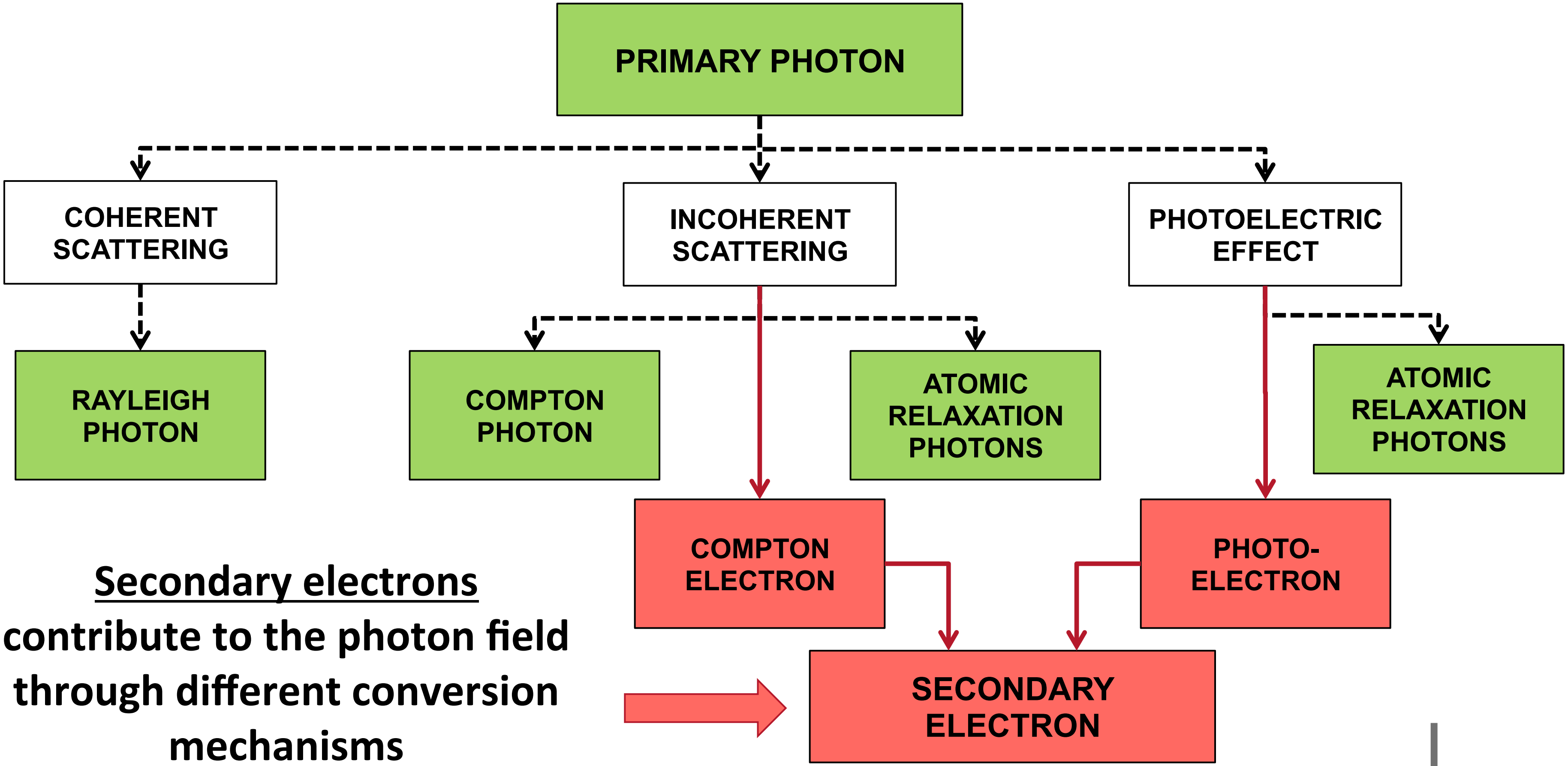
- Detector modification
  - Pulse pile-up.
  - Detector response function.

MULTIPLE SCATTERING IN X-RAY SPECTROMETRY

- X-rays penetrate deeply into the matter, and, in a thick medium, give place to a phenomenon known as multiple scattering (i.e, multiple collisions).
- Multiple scattering characterizes the radiation field in a thick medium
- Multiple scattering models describe the influence of the prevailing interactions in the x-ray regime to describe the radiation field.
- The emitted X-ray fluorescence spectrum is easily obtained from the radiation field, and is strongly modified by multiple scattering.
- Another important factor to characterize the radiation field is the effect of the polarization.

Scheme of X-ray interaction mechanisms

The full description of the radiation field requires the modeling of coupled photon-electron transport



Secondary electrons contribute to the photon field through different conversion mechanisms

Multiple scattering is usually described using the Boltzmann transport model

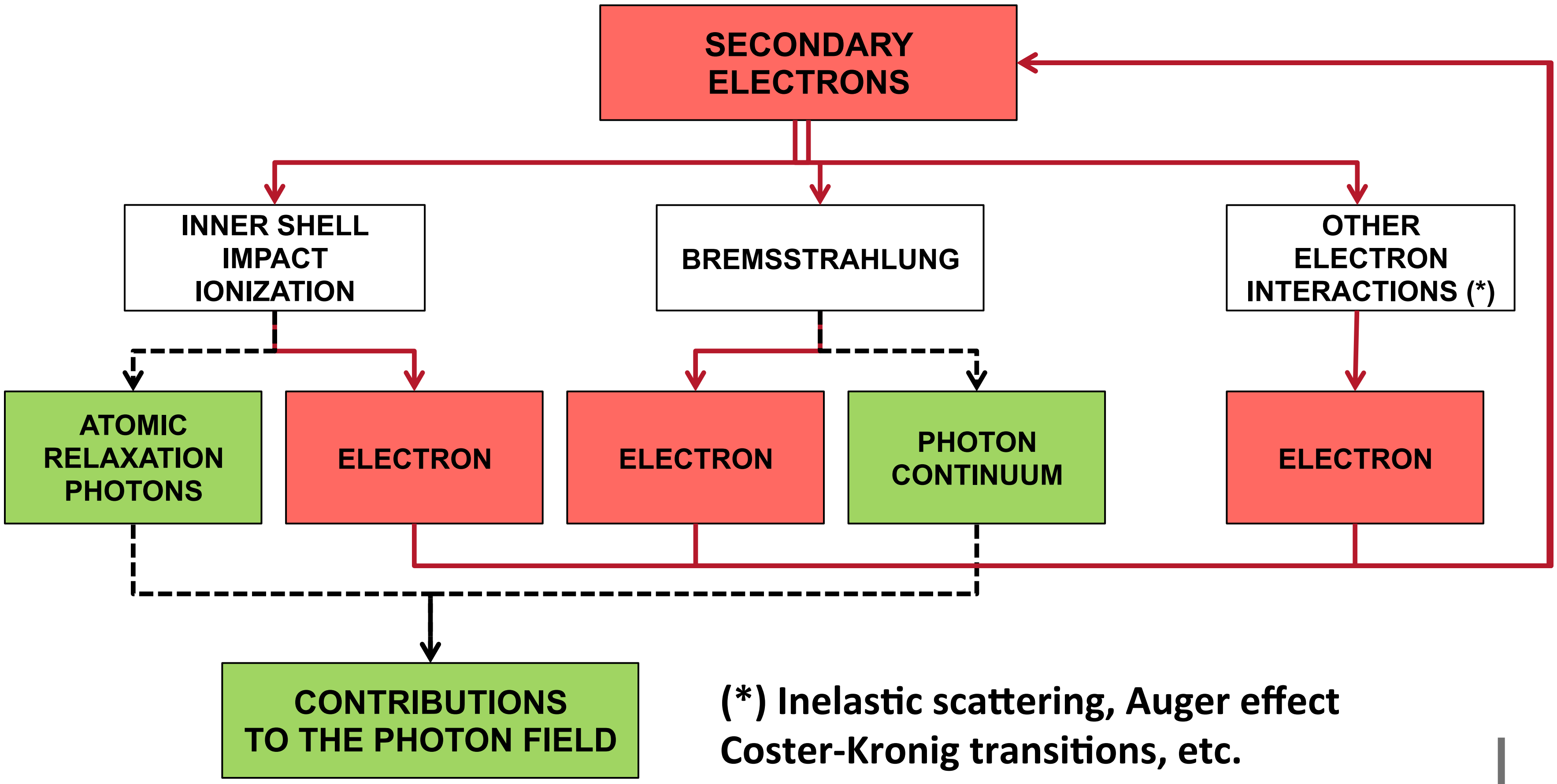
The photon interactions are depicted with the interaction kernels k_i

$$\eta \frac{\partial}{\partial z} f(z, \vec{\omega}, E) = -\mu(E) f(z, \vec{\omega}, E)$$
$$+ \sum_{i}^{all\ interactions} \int_0^{\infty} \left(\int_{4\pi} \cup (z) k_i \left(\vec{\omega}', E', \vec{\omega}, E \right) f\left(z, \vec{\omega}', E' \right) d\omega' \right) dE'$$
$$+ S(z, \vec{\omega}, E)$$

Not all the radiative contributions involved in an X-ray transport process are considered by the Boltzmann model and by the used interaction kernels k_i

X-ray production mechanisms from coupling terms

The full description of the radiation field requires the modeling of coupled photon-electron transport



(*) Inelastic scattering, Auger effect Coster-Kronig transitions, etc.

MS is better described using the modified Boltzmann transport model

The Boltzmann transport model has been recently modified to include the electron-photon contributions

$$\eta \frac{\partial}{\partial z} f^{p}(z,\omega,E) = -\mu^{p}(E) f^{p}(z,\omega,E) + \sum_{i}^{\text{all photon interactions}} \int_{0}^{\infty} \left(\int_{4\pi} \mathrm{U}(z) k_{i}^{p\to p}(\omega',E',\omega,E) f^{p}(z,\omega',E') d\omega' \right) dE' + \sum_{j}^{\text{all coupling terms}} \int_{0}^{\infty} \left(\int_{4\pi} \mathrm{U}(z) k_{j}^{e\to p}(\omega',E',\omega,E) f^{p}(z,\omega',E') d\omega' \right) dE' + S^{p}(z,\omega,E)$$

We need to include also the polarization WHY POLARIZATION?

Polarization state → **wave nature of photons**

By considering polarization we improve the model of photon diffusion

↓

a good approximation in many cases, but not for phenomena that are influenced by their wave properties

PHOTON DIFFUSION IS DESCRIBED BY A "VECTOR" TRANSPORT EQUATION (THE 1-D EQUATION IS SHOWN HERE)

$$\eta \frac{\partial}{\partial z} f^{p}(z,\omega,E) = -\mu^{p}(E) f^{p}(z,\omega,E) + \sum_{i}^{\text{all photon interactions}} \int_{0}^{\infty} \left(\int_{4\pi} U(z) H_{i}^{p \to p}(\omega',E',\omega,E) f^{p}(z,\omega',E')\, d\omega' \right) dE' + \sum_{j}^{\text{all coupling terms}} \int_{0}^{\infty} \left(\int_{4\pi} U(z) H_{j}^{e \to p}(\omega',E',\omega,E) f^{p}(z,\omega',E')\, d\omega' \right) dE' + S^{p}(z,\omega,E)$$

where

$$\overline{f} = \begin{bmatrix} I(z,\overline{\omega},\lambda) \\ Q(z,\overline{\omega},\lambda) \\ U(z,\overline{\omega},\lambda) \\ V(z,\overline{\omega},\lambda) \end{bmatrix}$$

VECTOR TRANSPORT EQUATION (CONT.)

where

$$\mathrm{H}^{(S)}(\vec{\varpi},\lambda,\vec{\varpi}',\lambda')=\mathrm{L}^{(S)}(\pi-\Psi)\mathrm{K}^{(S)}(\vec{\varpi},\lambda,\vec{\varpi}',\lambda')\mathrm{L}^{(S)}(-\Psi')$$

$H^{(S)}$ = kernel matrix in the meridian plane of reference

$K^{(S)}$ = scattering matrix in the scattering plane of reference

IMPORTANT PROPERTIES OF THE "VECTOR" TRANSPORT EQUATION

- Describes the evolution of the full polarization state (not only the intensity of the beam)
- Is linear (for the Stokes representation)
- Requires the simultaneous solution of the whole set of transport equations
- Cannot be transformed in a scalar equation !! (due to the coupling in the scattering term)

TWO RELEVANT ASPECTS

- A collision always changes the polarization state
- The angular distribution for scattered unpolarized and polarized photons is very different

MCSHAPE

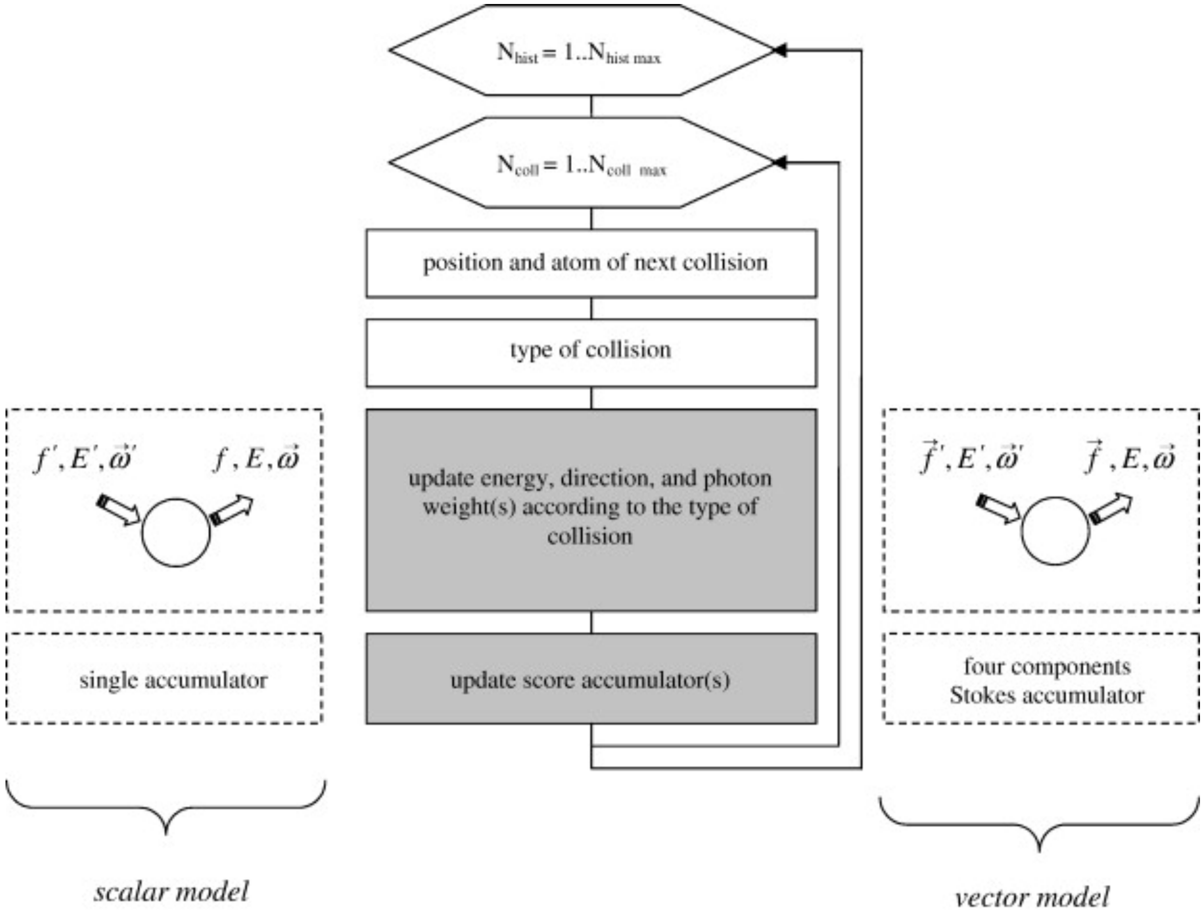
MCSHAPE is a Monte Carlo code developed at the University of Bologna which can simulate the diffusion of photons with arbitrary polarization state and has the unique feature of describing the evolution of the polarization state along the interactions with the atoms.

The adopted transport model is derived from the so called Boltzmann-Chandrasekhar 'vector' transport equation. The polarization state of the photons is described by using the Stokes parameters I, Q, U and V, having the dimension of intensities and containing the physical information about the polarization state.

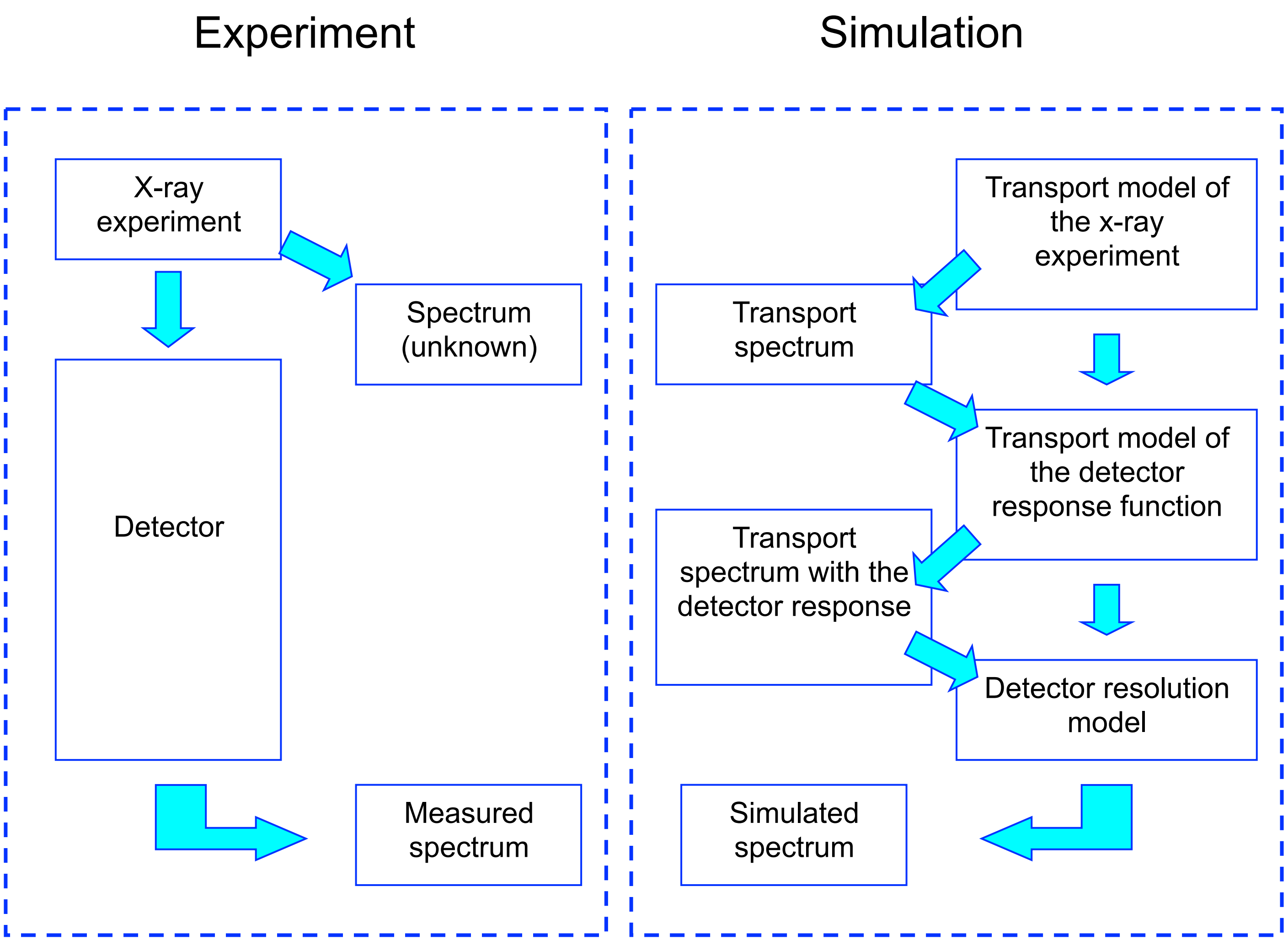
This code simulates the propagation in heterogeneous media of photons injected by either polarized (i.e., synchrotron) or unpolarized sources (x-ray tubes).

- Website: http://shape.ing.unibo.it

Differences between the computational structures of scalar and vector MC models



Schematic diagram of a simulation with MCSHAPE, compared with the experimental steps for a spectrum measurement.



Outline

- Detector modification
  - Pulse pile-up.
  - Detector response function.

Radiation Physics and Chemistry 104 (2014) 372–375

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem

Multi-shape pulse pile-up correction: The MCPPU code

Lorenzo Sabbatucci, Viviana Scot, Jorge E. Fernandez *

L. Sabbatucci, Viviana Scot, J.E. Fernandez: Multi shape pulse pile-up correction: the MCPPU code. Radiat. Phys. Chem. 104,(2014) 372-375

Radiation Physics and Chemistry 137 (2017) 12–17

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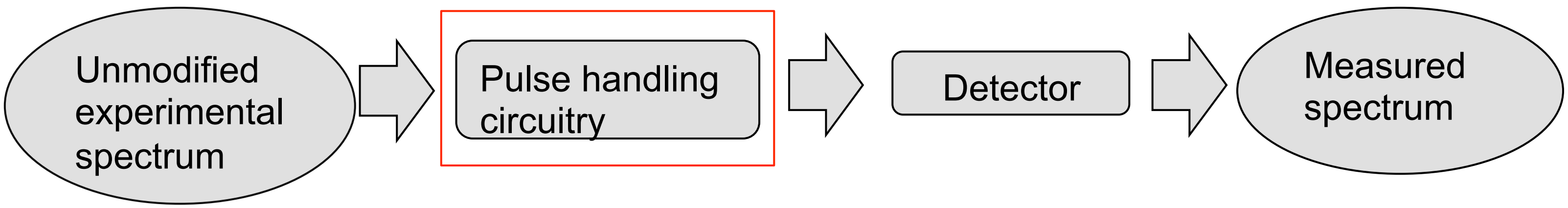
First principles pulse pile-up balance equation and fast deterministic solution

Lorenzo Sabbatucci [a], Jorge E. Fernández [a,b,*]

[a] Laboratory of Montecuccolino-DIN, Alma Mater Studiorum University of Bologna, via dei Colli 16, 40136 Bologna, Italy
[b] Alma Mater Studiorum Università di Bologna Representacion en la Republica Argentina, M. T. de Alvear 1149, Buenos Aires, Argentina

L. Sabbatucci, J.E. Fernandez: First principles Pulse Pile-Up balance equation and fast deterministic solution, Radiat. Phys. Chem. 137 (2017) 12-17

Pulse Pile-Up problem



- Radiation emission is a process randomly spaced in time. At high counts many pulses have time gap much smaller then their width, then pile-up effects occur.

Pulse pile-up has three consequences:

- Loss of counts: the sum of the pulses is detected instead of the separate pulses
- Piled-up pulses are distributed at wrong energies
- The whole spectrum is distorted since the lost pulses are not collected at the proper energies

- Modern detectors usually comprise built-in electronics which partially reduce pile-up effects either by limiting the pulse tail (i.e. Rise Time Discrimination, RTD) and/or by using additional rejection circuits.

Monte Carlo Approach: The code MCPPU

- MCPPU (Monte Carlo Pulse Pile-Up) is a general purpose code to perform post-processing pile-up correction on spectra obtained with different detectors.
- The code can correct pile-up distortions for spectra collected with or without electronic reduction circuitry.
- It is based on a modified version of the Monte Carlo algorithm developed originally by Gardner and Lee, Adv. X-ray Anal. 41 (1999) 941–950 and by Guo, Lee and Gardner, NIMA 531 (2004) 520-529.
- All orders of pile-up are taken into account.
- It allows the use of the pulse shape of the detector introduced by the user by means of an external text file.
- MCPPU automatically identify the dead time of the counting system to use in the pile-up recovery.
- MCPPU presents an user-friendly graphical interface.

L. Sabbatucci, V. Scot, J. E. Fernandez: Multi-shape pulse pile-up correction: the MCPPU code. Radiat. Phys. Chem. 104 (2014) 372–375.

Deterministic approach: code DRPPU

- It was derived a balance equation based on first principles assuming:
  1. an ideal MCA of infinitesimal energy bins width,
  2. only second order PPU,
  3. a rectangular pulse shape.

$$y(E) = [1 - 2\lambda\tau \exp(-\lambda\tau)]h(E) + N_t \lambda\tau \exp(-\lambda\tau) \int \bar{h}(E_1)\bar{h}(E - E_1) dE_1$$

- The solution of this equation is found using the following iterative strategy:

$$\omega(E) = -a x^{(i)}(E) + b \int x^{(i)}(E_1) x^{(i-1)}(E - E_1) dE_1$$

$$x^{(0)}(E) = \omega(E)$$

Where we define:

$$\omega(E) = \bar{y}(E) / N_t$$

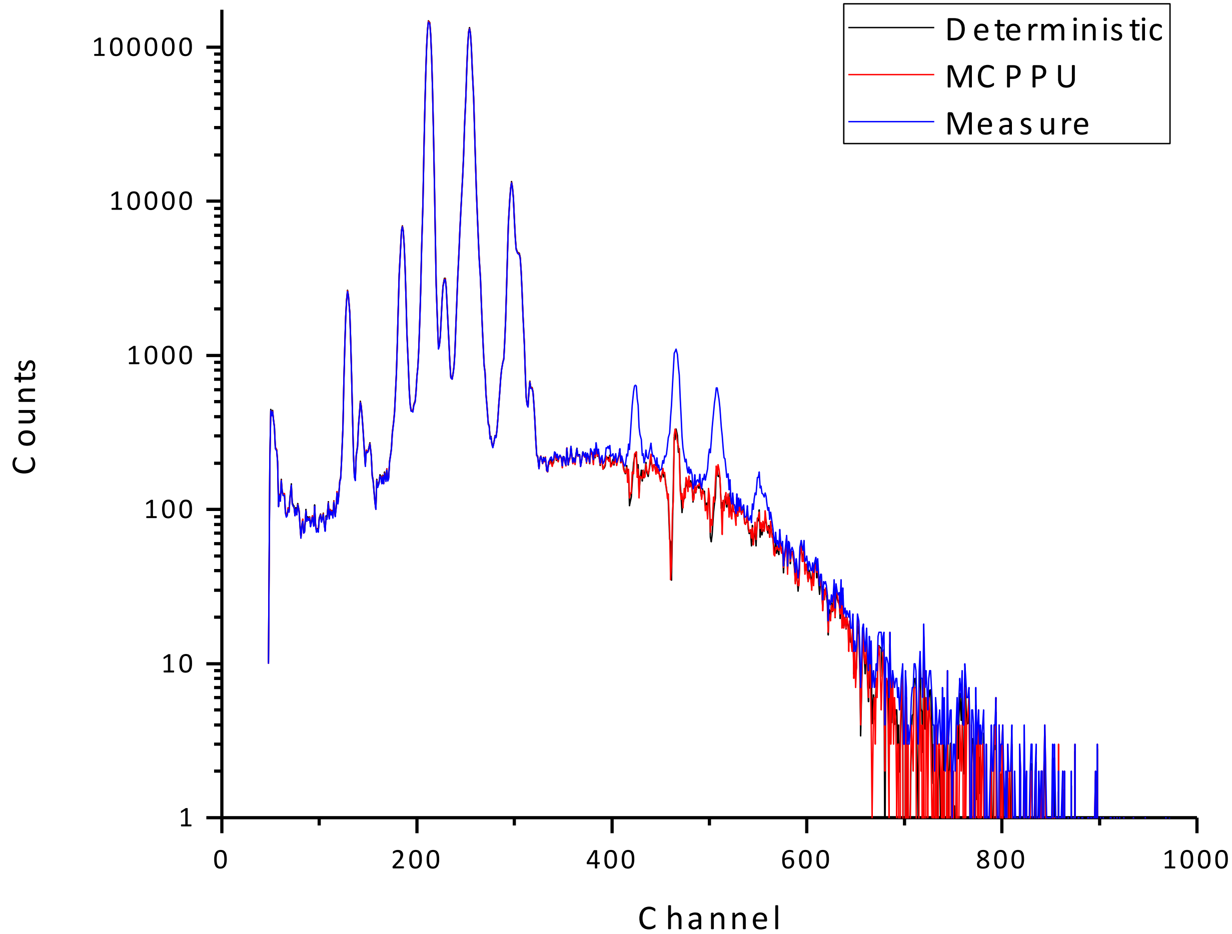
$$b = \lambda\tau \exp(-\lambda\tau)$$

$$a = 1 - 2b$$

$$N_t = \frac{\int y(E) dE}{1 - b}$$

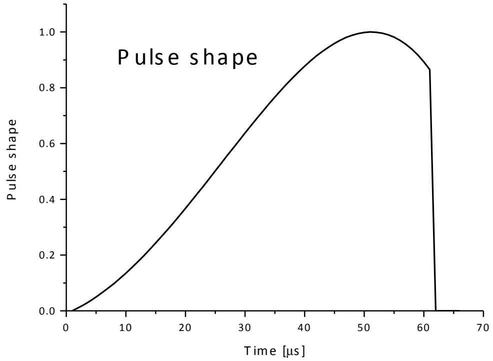
L. Sabbatucci, J.E. Fernandez: First principles Pulse Pile-Up balance equation and fast deterministic solution, Radiat. Phys. Chem. 137 (2017) 12-17

Pb sample



W X-ray tube operating at 35 kV
SSD Detector (Si): Amptek XR-100CR
Electronic rejection circuitry and RTD active

Live time: 600 s
Total counts: 2291631
Count rate: 3819.39
Dead time: 2.5e-6 s



Outline

- **Detector modification:**
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Radiation Physics and Chemistry 78 (2009) 882–887

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Simulation of the detector response function with the code MCSHAPE

Jorge E. Fernandez[a,b,*], Viviana Scot[a]

[a] Laboratory of Montecuccolino – DIENCA, Alma Mater Studiorum University of Bologna, via dei Colli, 16, I-40136 Bologna, Italy
[b] Istituto Nazionale di Fisica Nucleare (INFN), Italy

J.E. Fernandez, Viviana Scot: Simulation of the detector response function with the code MCSHAPE. Radiat. Phys. Chem. 78,(2009) 882-887

Research article

X-RAY SPECTROMETRY

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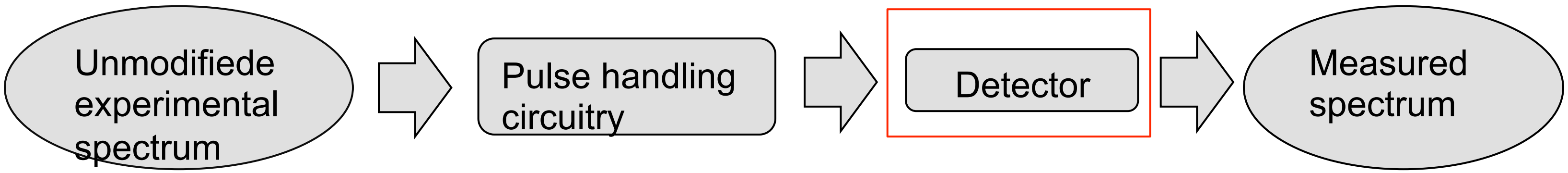
A modeling tool for detector resolution and incomplete charge collection†

Jorge E. Fernández, Viviana Scot and Lorenzo Sabbatucci*

J.E. Fernandez, Viviana Scot, L. Sabbatucci: A modeling tool for detector resolution and incomplete charge collection. X-ray Spectrometry 44 (2015) 177-182

Detector Response



(Detector influence on radiation measures)

The measured spectrum is given by the following convolution product:

$$I_{measured}(E) = \int R(E', E)\,\phi(E')\,I(E')\,dE'$$

Where

$R(E', E)$ is the response function

$\phi(E')$ is the detector efficiency

$I(E')$ is the original spectrum

Model of detector response

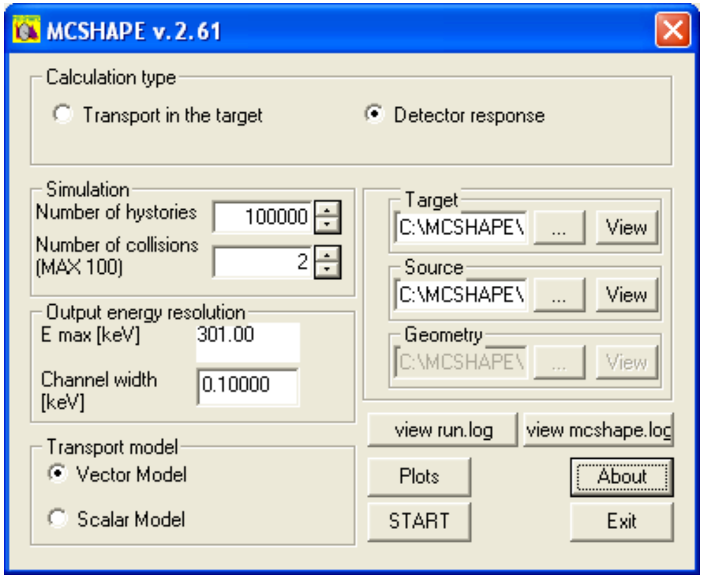
$$R(E_0, E) = \int Q(E'', E_0)\, G(E'', E)\, dE''$$

$Q(E'', E_0)$ is the energy deposition spectrum

$G(E'', E)$ is the detector resolution

Energy deposition spectrum

- Is built by computing the escape spectrum distribution
- In a first approximation its integral is normalized (really is not because of the Rayleigh scattering)
- It can be calculated using a MC code



MCSHAPE Computes the energy deposition spectrum

What is computed with MCSHAPE?

$$I_{measured}(E) = \int R(E',E)\,\varphi(E')\,I(E')\,dE'$$

$$= \int \left(\int Q(E'',E')\,G(E'',E)\,dE'' \right) \varphi(E')\,I(E')\,dE'$$

$$= \underbrace{\int \left(\underbrace{\int Q(E'',E')\,\varphi(E')\,I(E')\,dE'}_{\text{computed by MCSHAPE v2.61}} \right) G(E'',E)\,dE''}_{\text{computed by postprocessor RESOLUTION}}$$

J..E. Fernandez, V. Scot : Simulation of the detector response function with the code MCSHAPE, Radiation Physics and Chemistry 78 (2009) 882–887.

Detector resolution

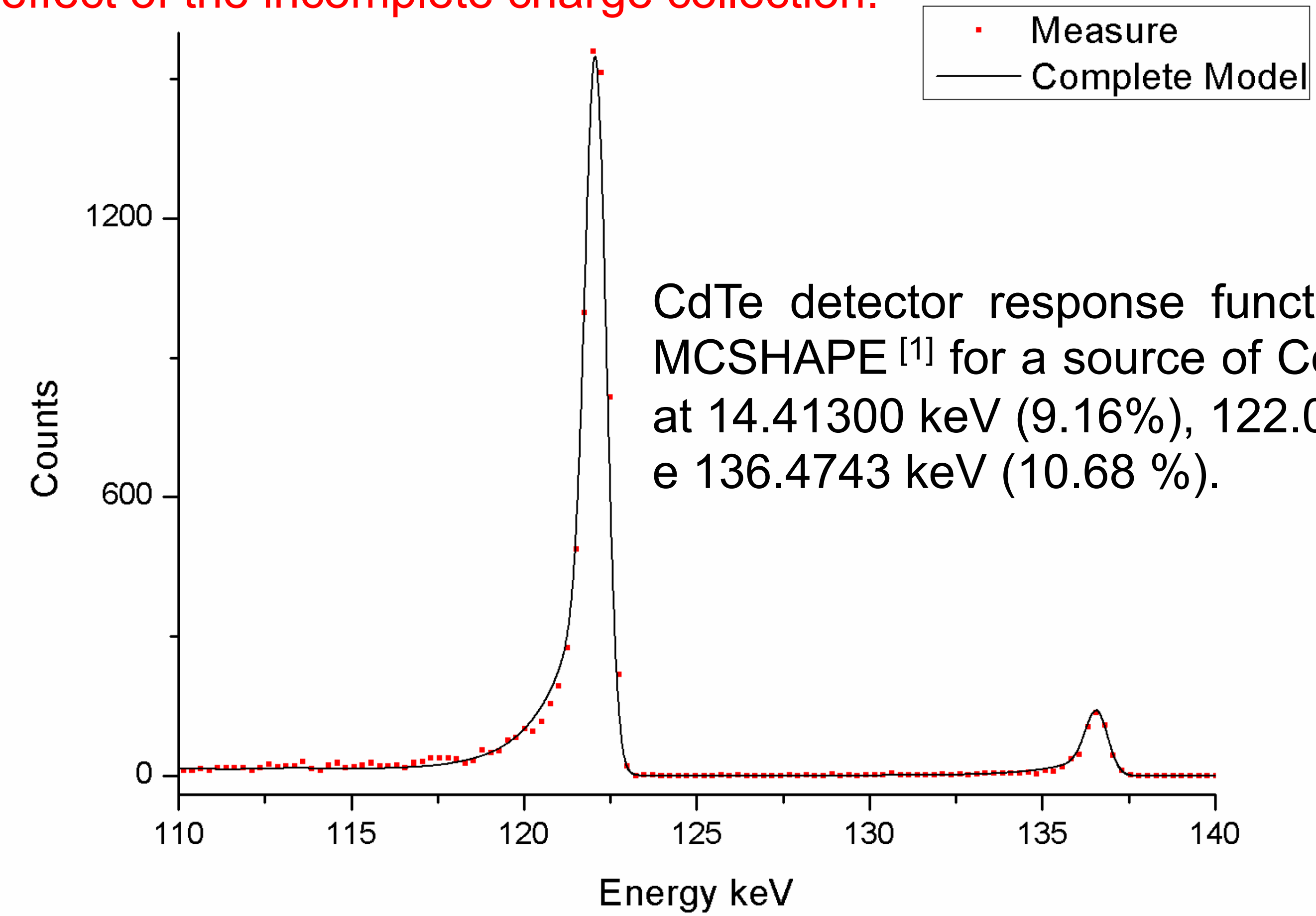
- In a first approximation it is described by a normalized Gaussian

$$G(E_0, E) = \frac{0.9395}{FWHM(E_0)} \exp\left\{-2.773 \frac{(E_0 - E)^2}{FWHM^2(E_0)}\right\}$$

- the FWHM is a function of energy

RESOLUTION: CdTe response function

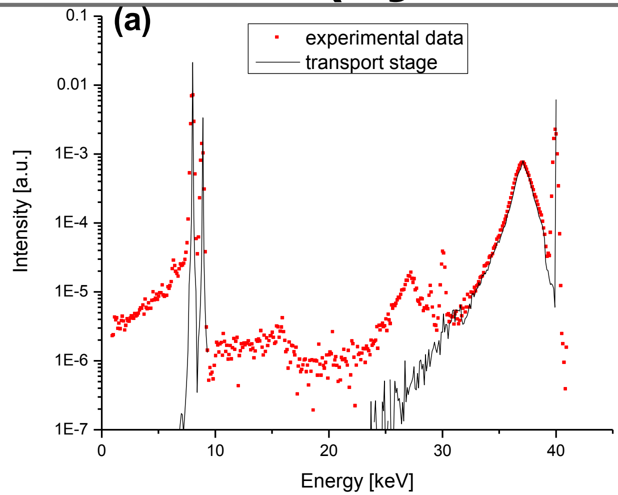
The new version of RESOLUTION allows also, for a solid state detector, to introduce the effect of the incomplete charge collection.



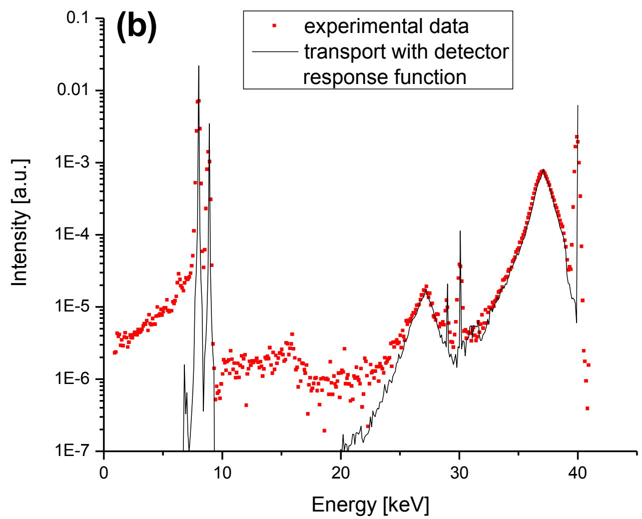
CdTe detector response function computed with MCSHAPE [1] for a source of Co57, with emission at 14.41300 keV (9.16%), 122.0614 keV (85.60 %) e 136.4743 keV (10.68 %).

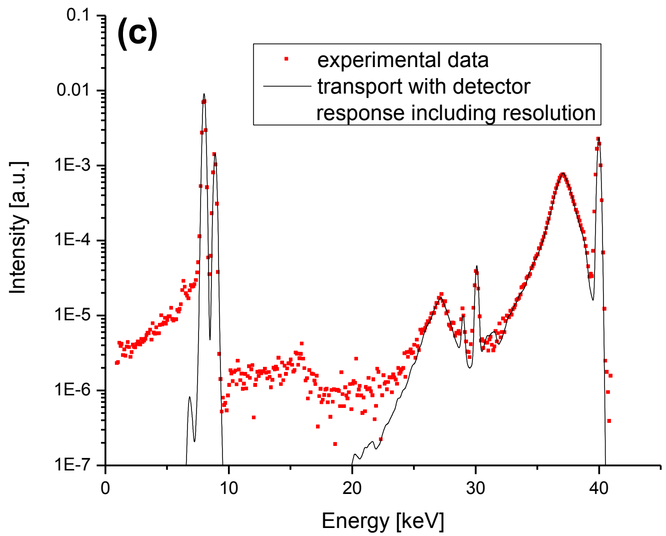
J. E. Fernández, V. Scot, L. Sabbatucci: A modeling tool for detector resolution and incomplete charge collection, X-ray Spectrom. 44 (2015) 177-182.

Comparison with experimental data (synchrotron experiment)



- Sample: Cu
- Energy: 40 keV
- Linearly polarized source with polarization degree P= 0.885
- Scattering angle: 90°

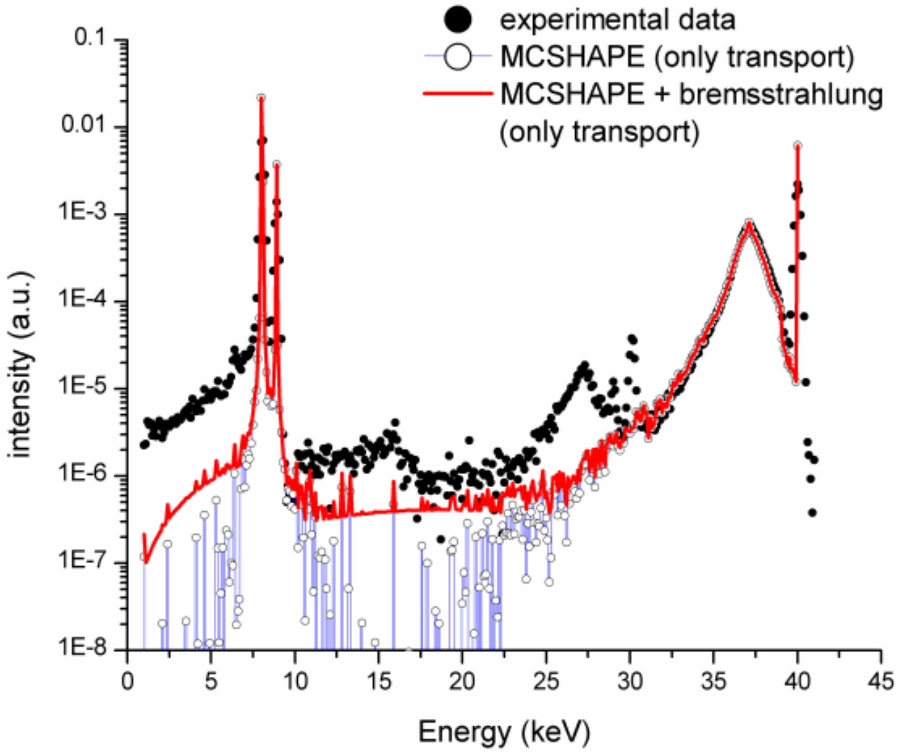


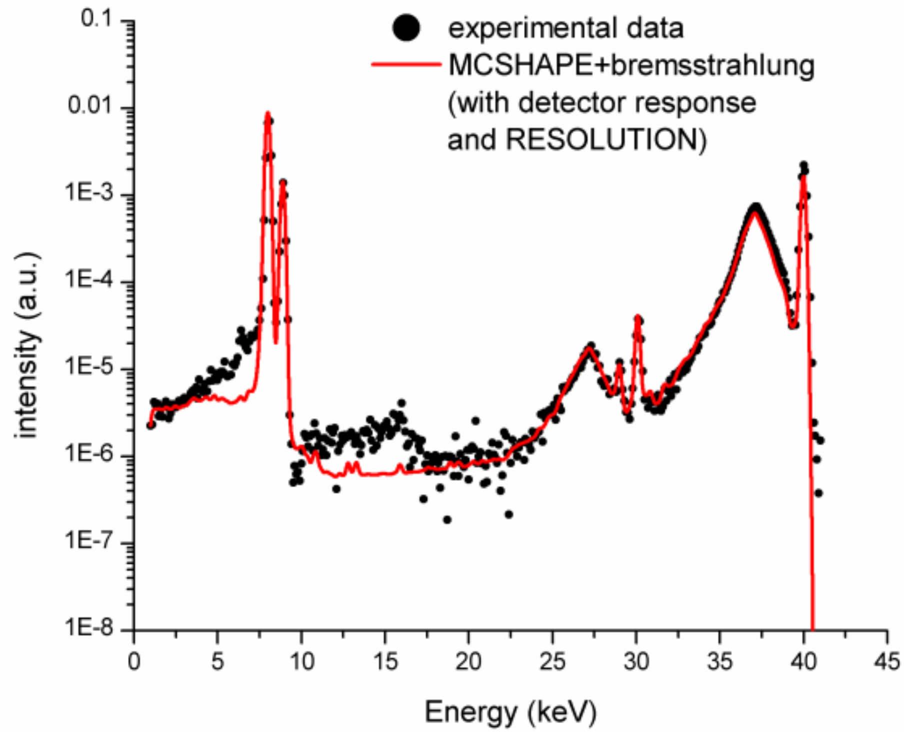


[1] Fernandez, J.E. and Scot, V. (2009), Simulation of the detector response function with the code MCSHAPE. Rad. Phys. Chem. 78(10):882-887

Comparison with experimental data (synchrotron experiment)

Comparison between the experimental data and the simulation of the transport in the target performed with the MCSHAPE[1] with and without bremsstrahlung contribution





ALMA MATER STUDIORUM
UNIVERSITÀ DI BOLOGNA

Thank you for your attention!

jorge.fernandez@unibo.it