AGH UNIVERSITY OF SCIENCE AND TECHNOLOGY

Application of gaseous detectors for full-field EDXRF imaging of works of art

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Presentation outline

- Introduction to gaseous detectors
- Position-sensitive gaseous detectors
- The idea of full field EDXRF imaging - optics
- The gaseous detector based FF-EDXRF imaging systems
- Conclusions

Signal generation in gas detectors

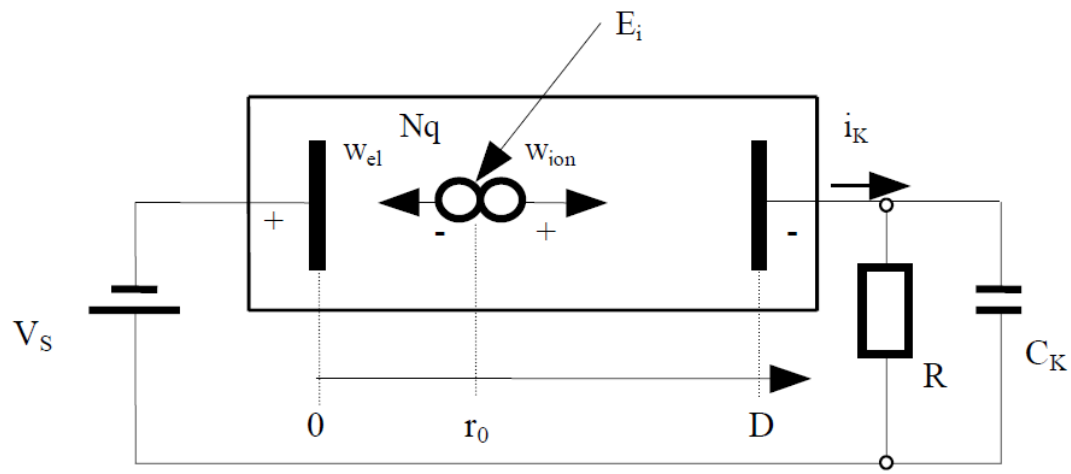
Simplest gas detector – flat ionisation chamber

1) Photoelectric absorption of X ray photon in gas medium
2) Photoelectron and Auger electron ionize gas

$$N = \frac{E}{W} \quad , W \approx 25\ eV/pair$$

$$N \approx 200\ (for\ 5\ keV)$$

3) Ions and electrons **drifts** towards electrodes in electric field



Shockley – Ramo theorem

Drift of charge induce current:

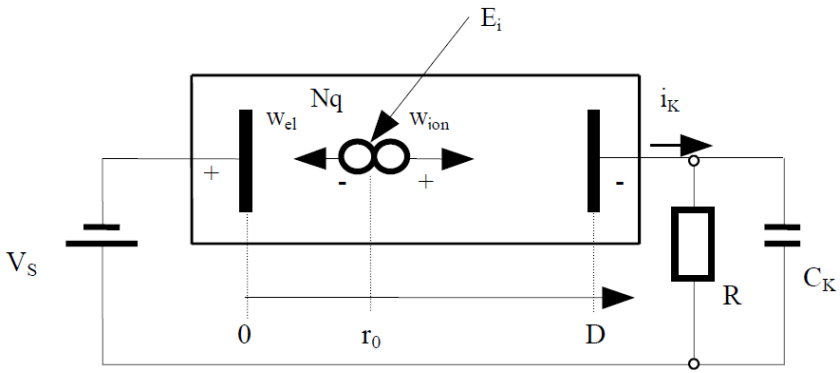
$$i_k(t) = Nqv_k(t) \cdot \Psi(\vec{r}) \qquad v_k(t) = \mu_k E(\vec{r}(t))$$

mobility:

$$\mu_{electron} \approx cm/\mu s$$

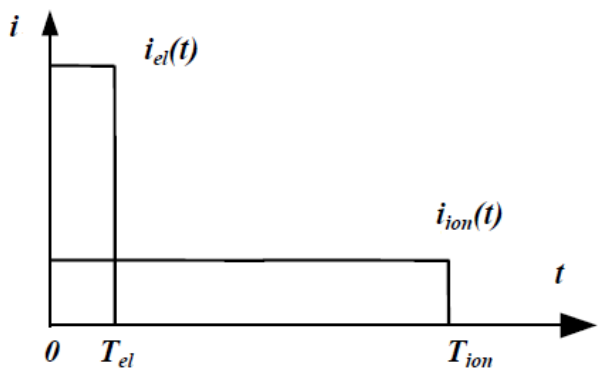
$$\mu_{ion} \approx cm/ms$$

For flat ionisation chamber:



Charge collection time:

$$T_{ion} \gg T_{electron}$$



Very long tail pulse that depends on:
-interaction position
-detector size

Proportional counter – avalanche multiplication

higher HV

↓

stronger electric field

↓

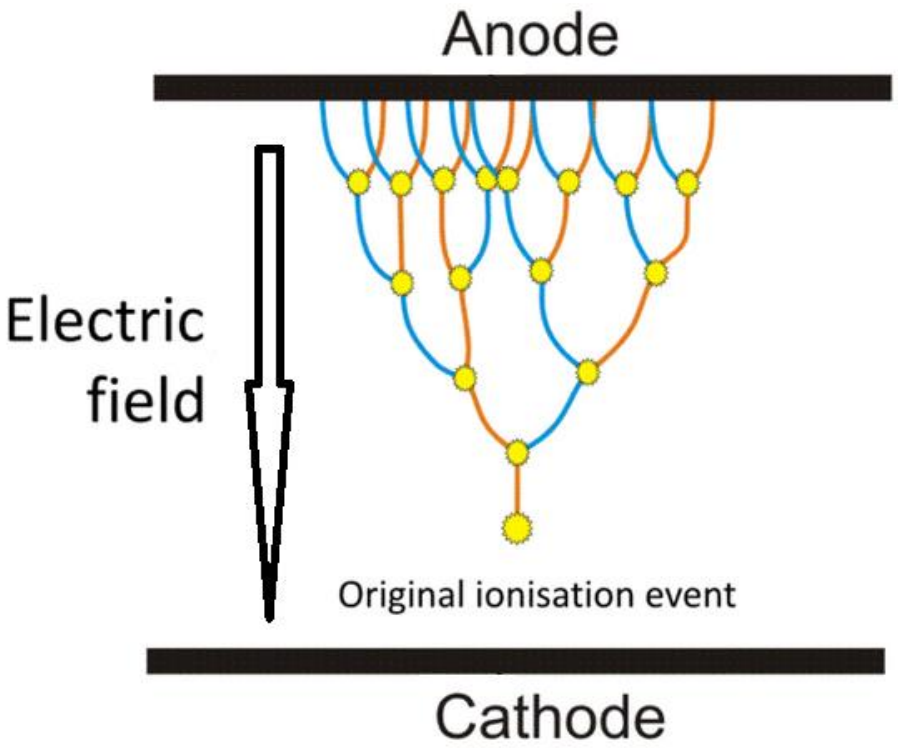
greater energy of drifting electrons / ions

↓

electrons causes secondary ionisation

↓

avalanche multiplication – gas gain



typical gain: 10^2 - 10^6

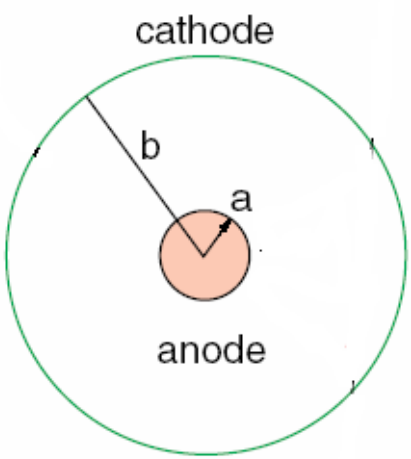
huge number of ion pairs – high current pulse
still dependant on energy of photon

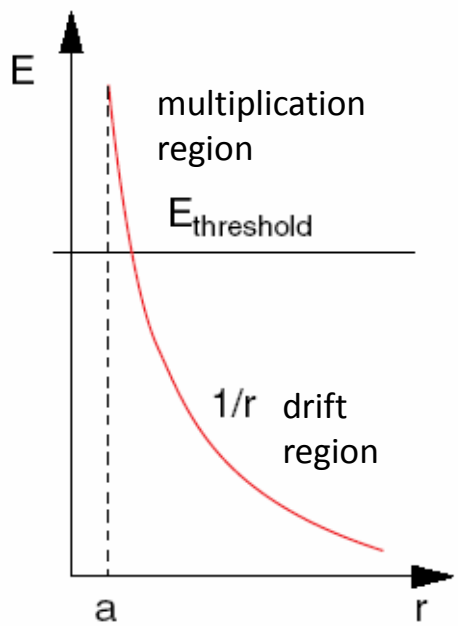
Proportional counter

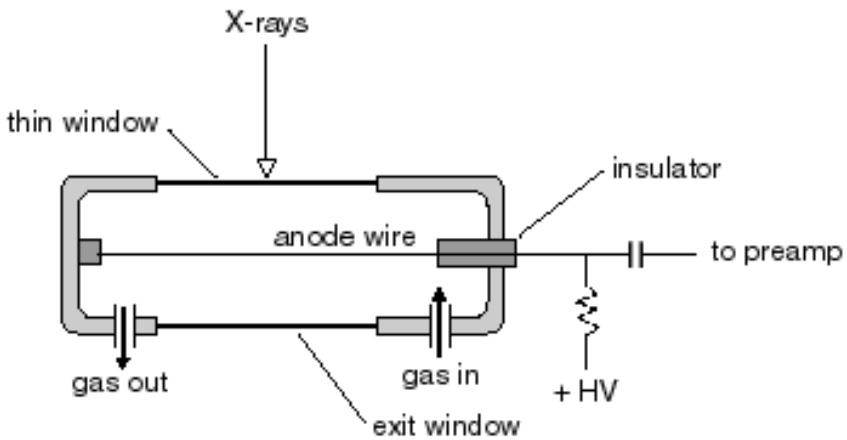
Typical cylindrical geometry:

-strong electric field (~10^6 V/m) near very thin anode (<100µm) without very high voltage

-the multiplication occurs near the anode wire







Space-charge effects

- avalanche multiplication create ion cloud around the anode
- electrons are rapidly collected (~1ns)
- cloud of positive ions create space charge that moves slowly towards cathode

- space charge attenuate electric field and thus decreases the gas multiplication

nonlinearity, gain fluctuations

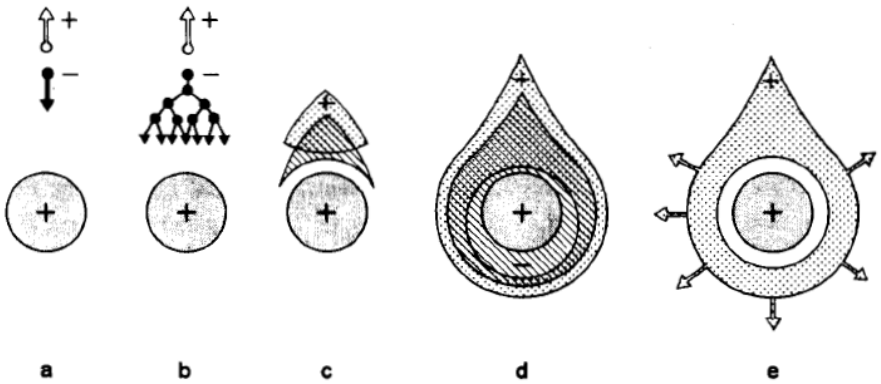


Fig. 49 Time development of an avalanche in a proportional counter[30]. A single primary electron proceeds towards the anode, in regions of increasingly high fields, experiencing ionizing collisions; due to the lateral diffusion, a drop-like avalanche, surrounding the wire, develops. Electrons are collected in a very short time (1 nsec or so) and a cloud of positive ions is left, slowly migrating towards the cathode.

F. Sauli, CERN-77-09, [G. Charpak, Filet a particules, Découverte (février 1972)]

stronger field – stronger multiplication – more ions – more space charge effect

Geiger-Müller counter

at very high electric field (high voltages):

Ionisation + excitation of gas molecules/atoms

deexcitaiton of molecules – creation of UV photons

absorption of UV photons – creation of new ion pairs

new avalaneches and so on…

Until:

ALL gas around anode wire (~10^{-5}s) is ionised

the process is stopped by space charge effect

(electric field becomes weak – charge multiplication impossible)

can be reduced
by addition of quenching
gas i.e. CO_2, CH_4

very strong impulse
but
energy information lost

Gaseous vs solid state detectors

Energy resolution

fluctuations of N and gas gain gives $\frac{\Delta E}{E} \approx 15\%$

Detection efficiency

low density medium – limited quantum efficiency at high energy

BUT – they are cheap and does not requres sensitive signal processing electronics

Position sensitive gaseous detectors

- Multi-Wire Proportional Chamber (MWPC) - 1968

Micro Pattern Gas Detectors (MPGD):

- Micro Strip Gas Chamber (MSGC) - 1988
- MicroMEGAS – 1996
- **Gas Electron Multiplier (GEM) – 1997**
  - **THGEM, TGEM, MHSP, THCOBRA**

all of them developed for detection of charged particles in high energy physics experiments !

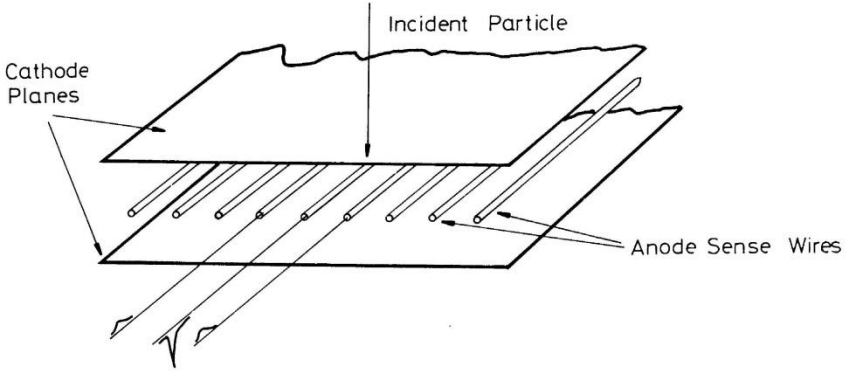
Multi Wire Proportional Chamber

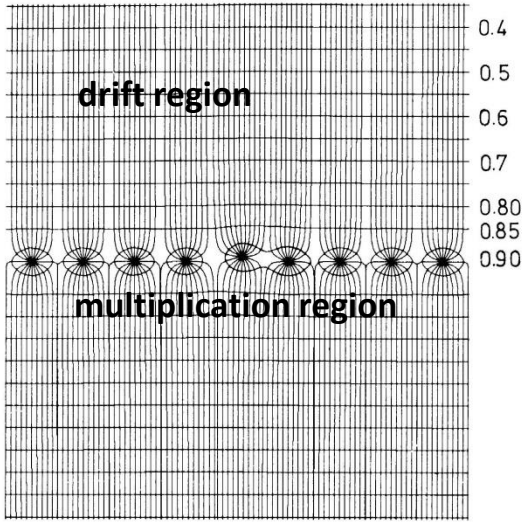
G. Charpak – 1968 (Nobel prize 1992)

Typical dimensions

- Thickness : 10 mm
- Anode wires separation: 1 mm
- Anode wires lenght : 20 mm
- Spatial resolution limit : 300 µm

  (pitch / √12)



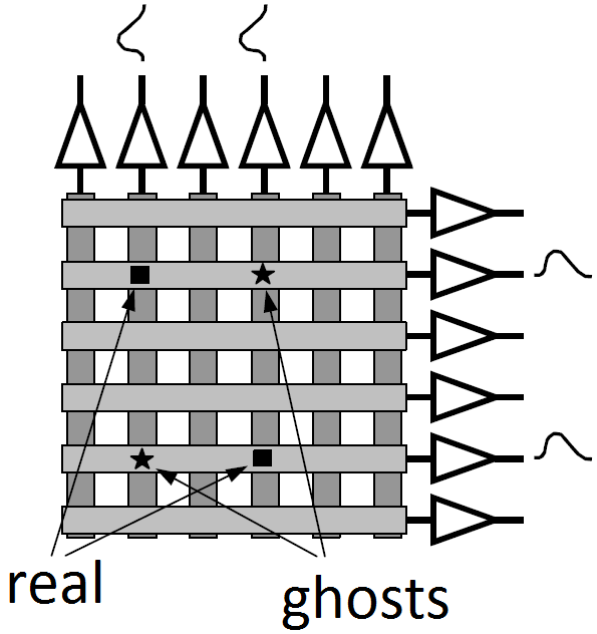


G. Charpak, D. Rahm, H. Steiner, NIM 80, Issue 1, 1970, 13-34

Multi Wire Proportional Chamber

Secondary coordinate

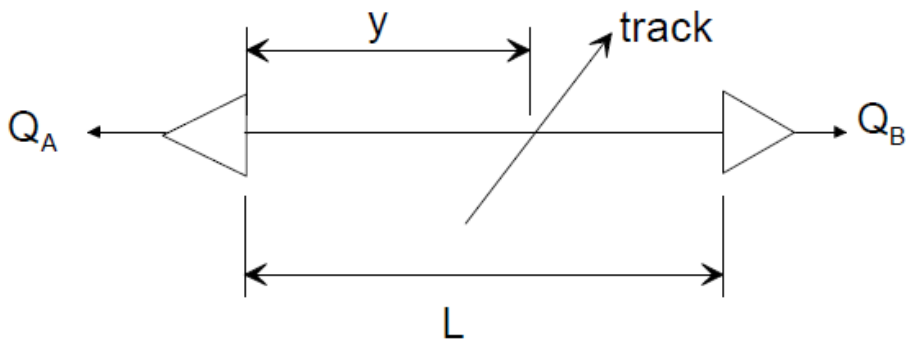
- Crossed wire planes



- Charge division (resistive wires)

$$\frac{y}{L} = \frac{Q_B}{Q_B + Q_A}$$

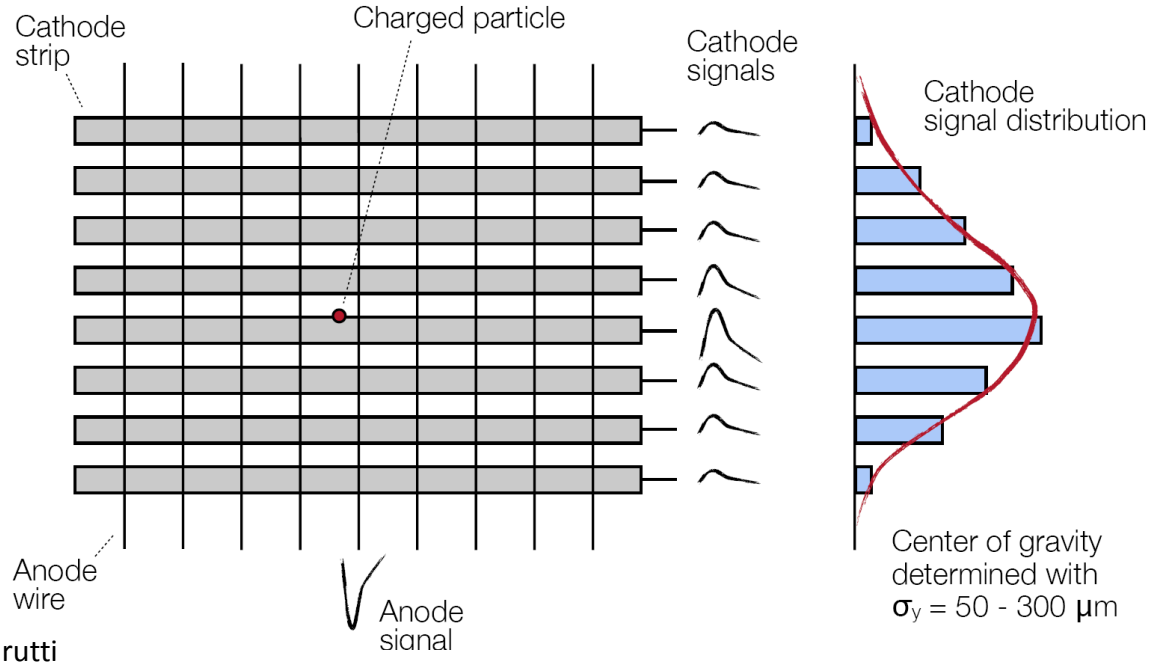
$$\sigma\left(\frac{y}{L}\right) \approx 0.4\%$$



Multi Wire Proportional Chamber

Secondary coordinate

- Segmented cathode – second coordinate given by ions
  - Ions not only drift but also diffuse – but center of gravity method!



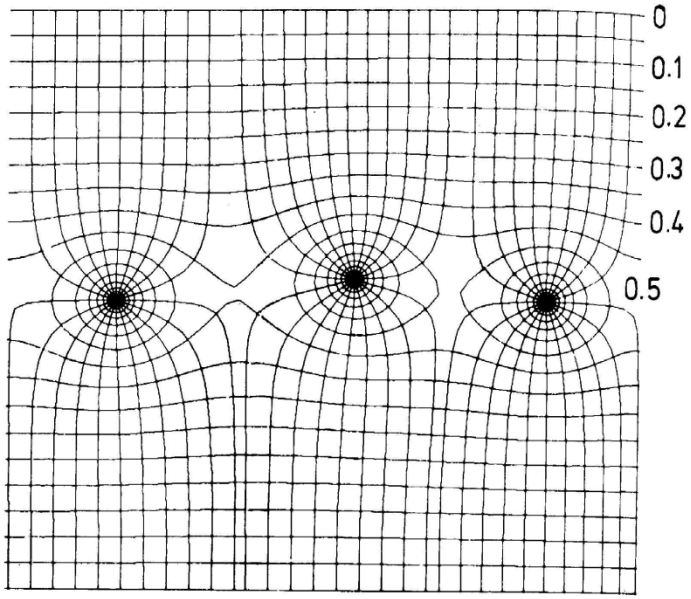
http://www.desy.de/~garutti

Multi Wire Proportional Chamber

Problems:

- Long ion path (few mm) = long pulses = low rate (10^4 Hz/mm^2)
- Electrostatic repulsion –

  anode displacement effect that reduces field quality
- Space charge effect



G. Charpak, D. Rahm, H. Steiner, NIM 80, Issue 1, 1970, 13-34

Micro Strip Gas Chamber

A.Oed – 1988

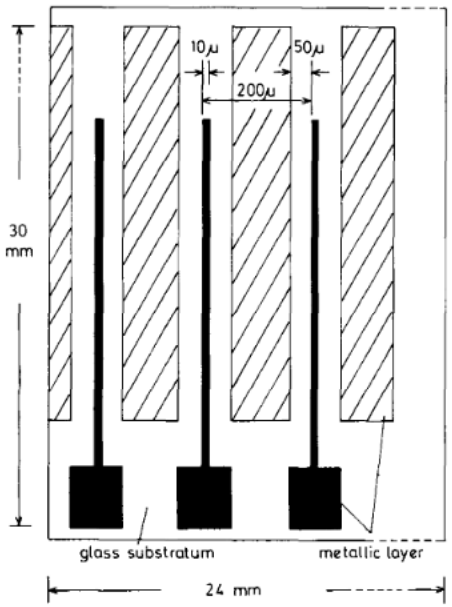


Fig. 1. Dimensions of the microstrip plate (MS-plate) used for the measurements. The chromium layer on the glass substrate is 300 nm in thickness.

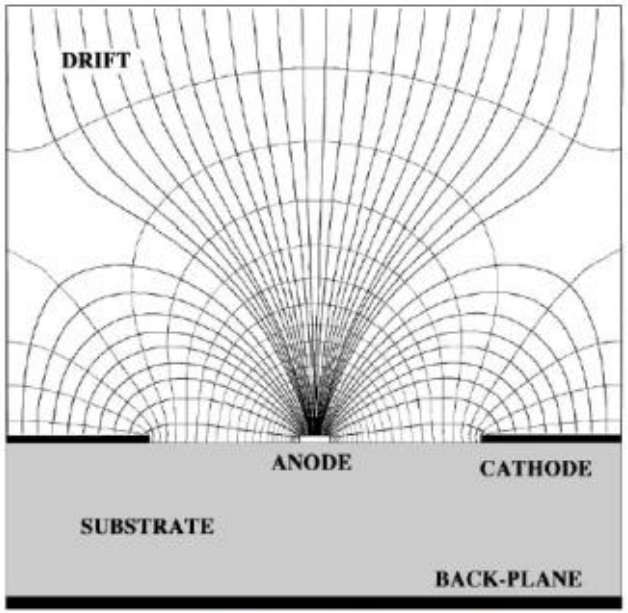


Figure 2 Equipotentials and field lines in the microstrip chamber, computed close to the substrate. The back-plane potential has been selected to prevent field lines entering the dielectric.

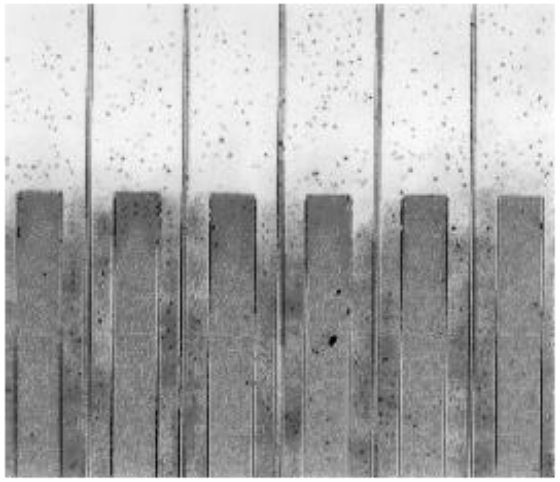


Figure 1 Close view of one of the first microstrip plates developed by Oed at the Institut Laue-Langevin. On an insulating substrate, thin metallic anode strips alternate with wider cathodes; the pitch is 200 μm.

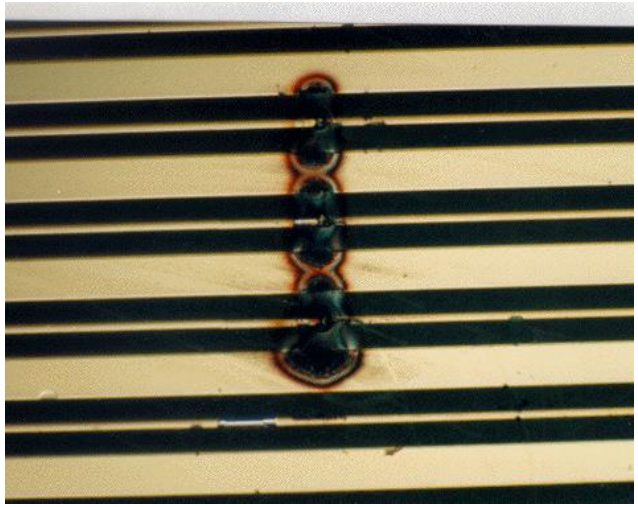
usually Cr/Au strips on glass substrate (300 µm) photolitography

F,Sauli, A. Sharma, Annu. Rev. Nucl. Part. Sci. 1999. 49:341–88

second coordinate – segmenting of bottom side of the insulator

Micro Strip Gas Chamber

- Large area 100 X 100 mm^2
- Small distance between strips – spatial resolution ~30 µm
- Short path to travel for ions – short pulses – high speed (10^6 Hz/mm^2)
- Very high field – sparks (aging or damage)
- Charging of substrate material – gain fluctuations



https://gdd.web.cern.ch/GDD/msgc.htmL

MicroMEGAS – Micro MEsh GAseous Structure

I. Giomatris - 1996

- Small gap – high homogenous field
- Fast movement of ions (~100 ns)
- Very high count rates (10^7 Hz/cm^2)
- Small space charge effect
- Resolution up to 60 µm
- Possible sparks between strips and mesh

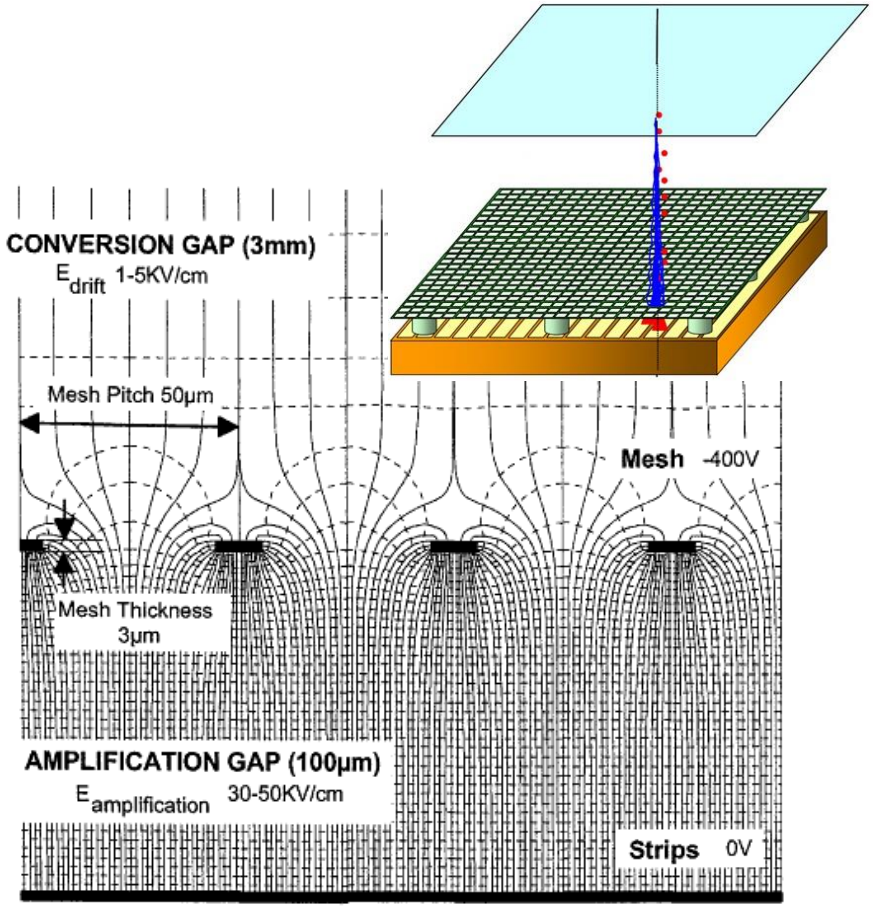
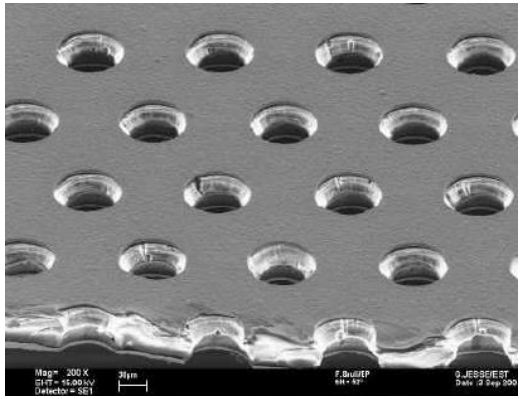


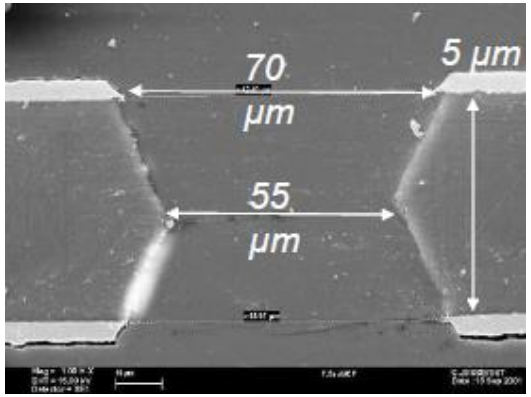
Fig. 1. Micromegas electric field map.

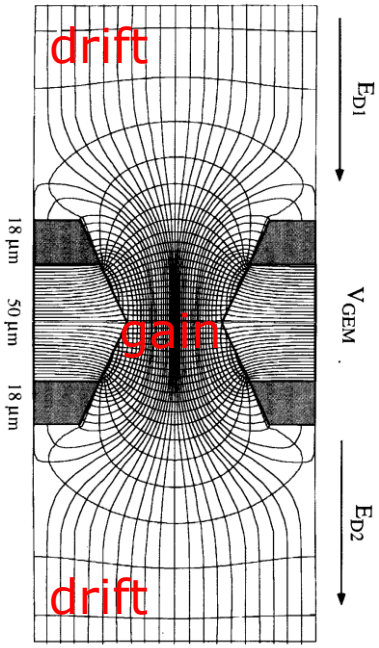
Gas Electron Multiplier

F. Sauli – 1997

- Cu electrodes separated with kapton (litography + etching)
- 100 holes / mm^2
- typical pitch ~140 µm
- typical voltage 350 – 500 V – E field ~50 kV/cm
- avalanche multiplication inside holes (10^2)







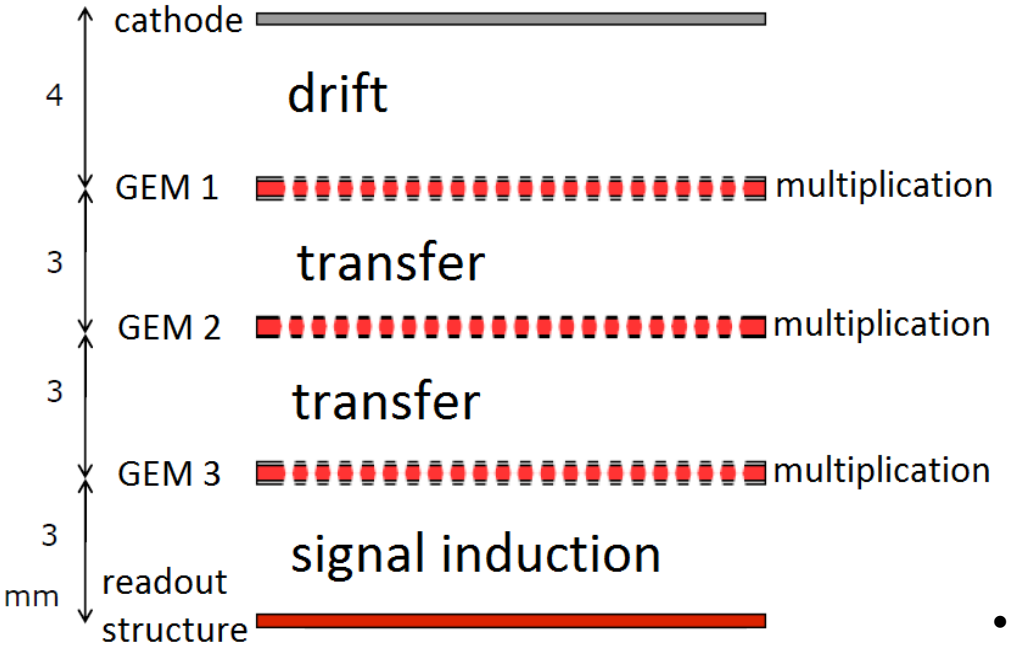
ThickGEM

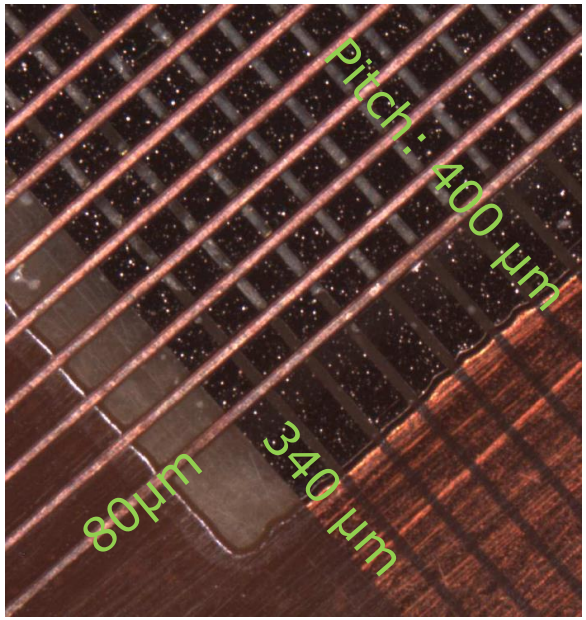
- 400 µm kevlar / fiberglass
- 0.3 mm drilled holes / 0.7 mm pitch
- 50 µm Cu electrodes
- voltage up to 2 kV – high gain (10^5)
- <u>worse spatial resolution</u>

F. Sauli, Nucl. Instr. and Meth. A386(1997)531

Triple GEM detector

Single GEM gives small gain (up to 10^2) – solution: use several GEM foils





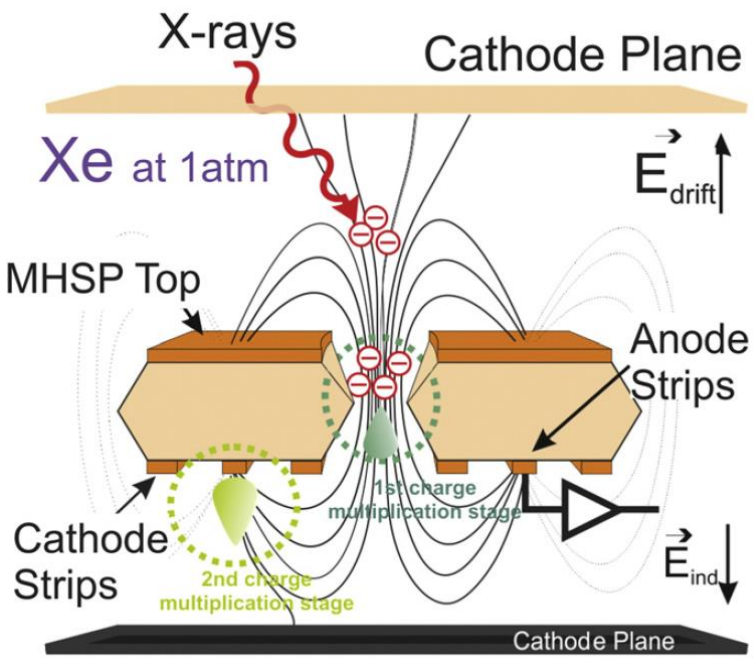
2D readout – crossed electrodes

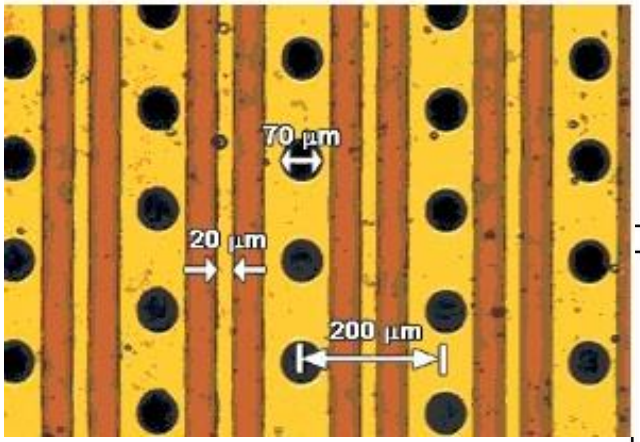
- ion movement negligible – very fast electron signal (<40 ns)
- count rate up to 10^6 Hz/mm^2
- resolution < 80 µm

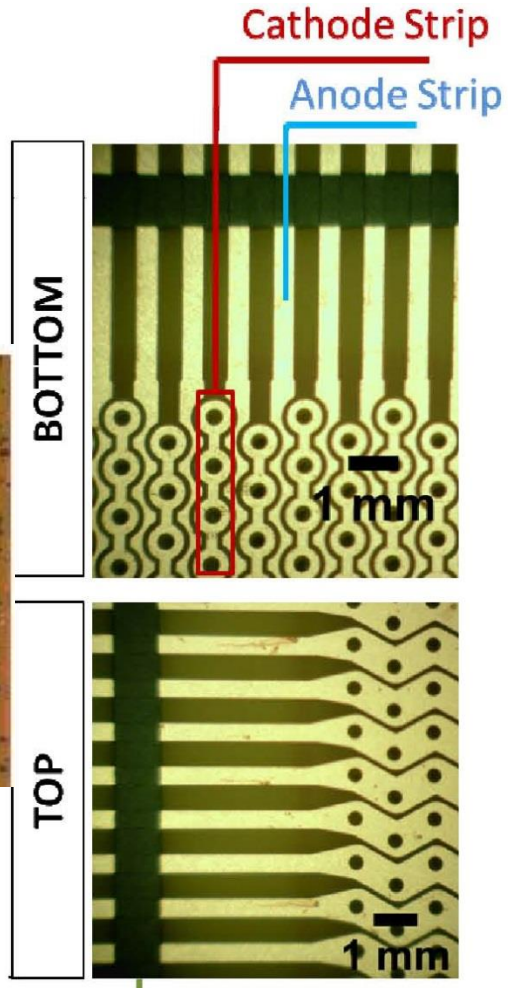
Multi Hole Strip Plate, ThCOBRA

Combination of GEM and MSGC = GEM with segmented electrodes

Secondary multiplication – high gain with one GEM-like structure







MHSP based on thin GEM, ThCOBRA based on ThickGEM

A.L.M. Silva et al. / Spectrochimica Acta Part B 86 (2013) 115–122

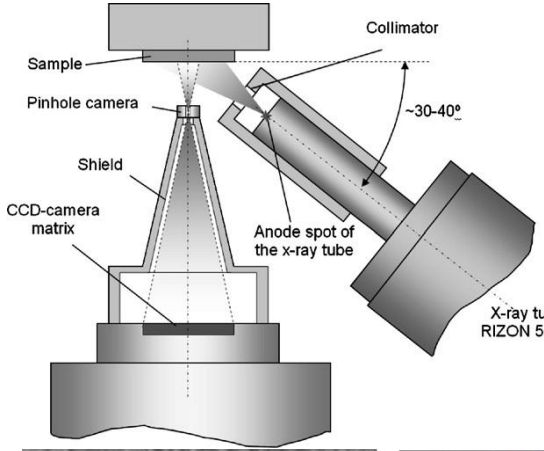
Position sensitive gaseous detectors - summary

- **Large area**
- **Low cost**
- **High count rate (10^6 Hz/mm^2)**
- **Resolution up to 60µm**
- **High amplitude of output signal (avalanche multiplication)**
- **Moderate energy resolution**
- **Time/spatial gain fluctuations (easy to compensate)**
- **Gas flow required**

Full field EDXRF imaging

- FF-EDXRF systems based on silicon detectors
- Pinhole optics
- FF-EDXRF systems based on gaseous detectors
- Conclusions

Full field EDXRF imaging – early examples



CCD
25.9x27.5 mm^2
1242x1152px
res: ~400 eV

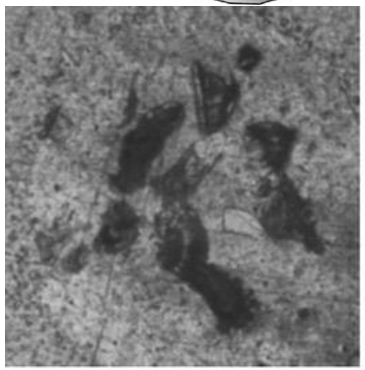
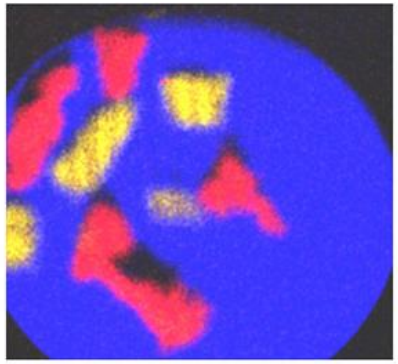


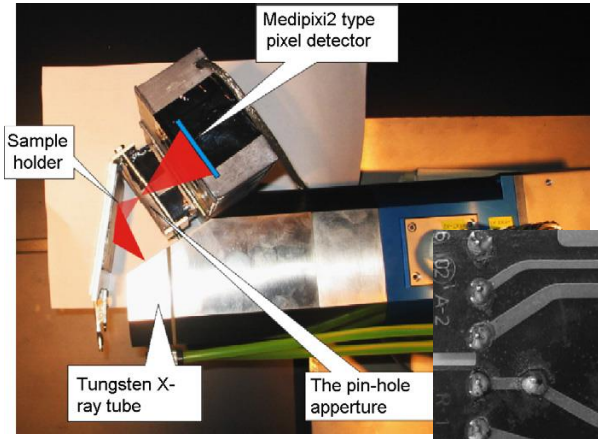
Photo of the sample in the optical range



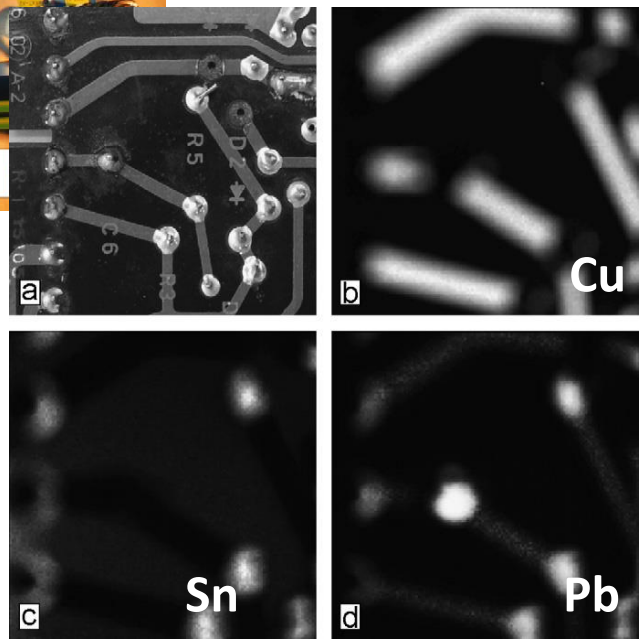
Resulting spectral-selective image of the sample surface

pure metallic Ti/Cu/Ge sample

M.G. Vasin et al. Spectrochimica Acta Part B 62 (2007) 648–653



hybrid-pixel
14x14 mm^2
256x256px
res: 3 – 5 keV



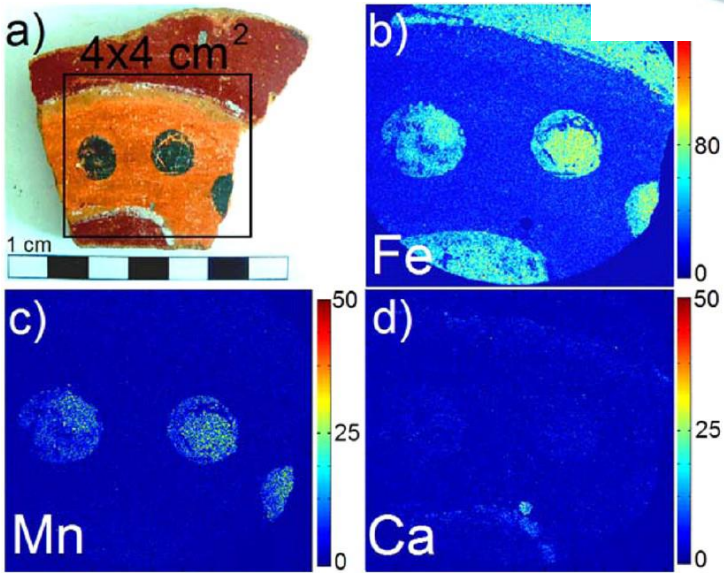
printed circuit board

V. Tichy et al. NIMA 591 (2008) 67–70

FF-EDXRF imaging of art works (a short reminder)

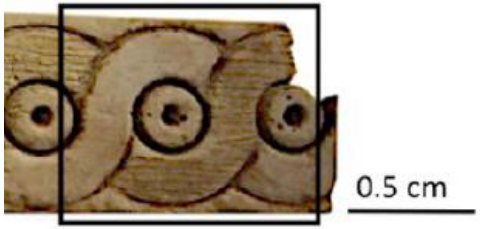
CCD based FF systems in CH investigation

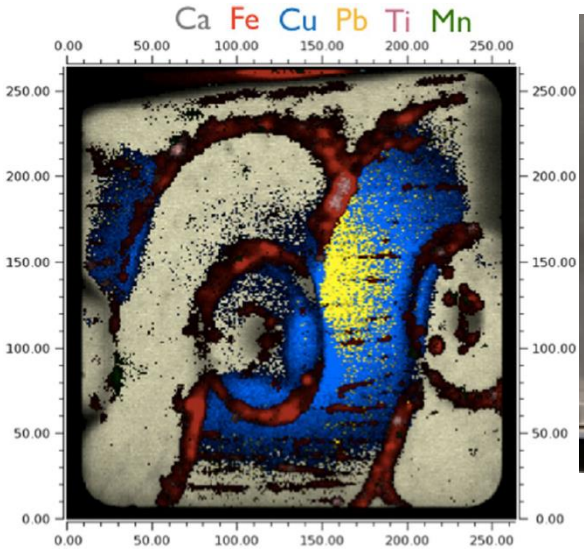
BI-DD-CCD
13x13 mm^2 1024x1024 px
FOV: 2.5x2.5mm^2 - 4x4 cm^2



Polychrome potery from Nasca (V A.D.)
5000s, 50µm pinhole, 170 µm resolution

F.P. Romano et al. Spectrochimica Acta Part B 86 (2013) 60–65
F.P. Romano et al. Anal. Chem. 2014, 86, 10892−10899



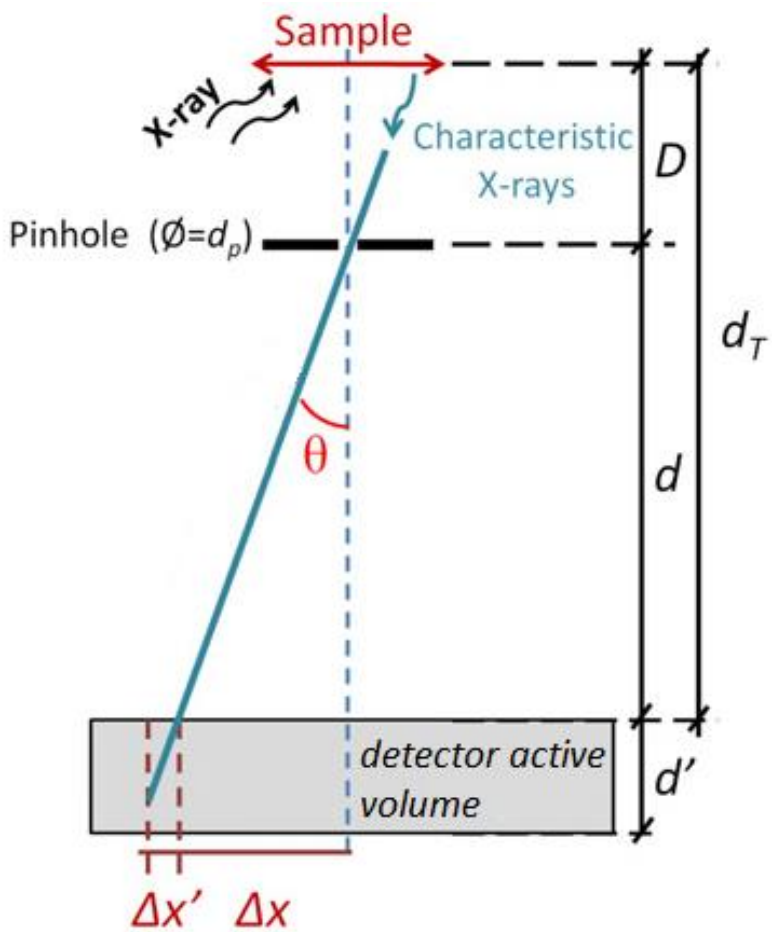


pn-CCD
12.7x12.7 mm^2
264x264 px
M: 1-6

polychrome wave frieze (VIII B.C.) ivory
40 – 100 min, PC optics, 48 µm resoluton

I. Reiche et al. Anal. Chem. 2013, 85, 5857−5866

Full field EDXRF imaging – pinhole optics



Magnification: $$M = \frac{d}{D}$$

Resolution: $$\sigma_{system} = \sqrt{{\sigma_{pinhole}}^2 + \frac{{\sigma_{det}}^2}{M^2} + {\sigma_{\Delta x}}^2}$$

$$\sigma_{pinhole} = \frac{d_p}{\sqrt{8\ln(2)}}\left(1 + \frac{1}{M}\right)$$

$$\sigma_{det} = \frac{p}{\sqrt{12}} \qquad \sigma_{\Delta x} = \frac{d'\Delta x/d_T}{\sqrt{8\ln(2)}M}\left(1 + \frac{1}{M}\right)$$

Sensitivity:

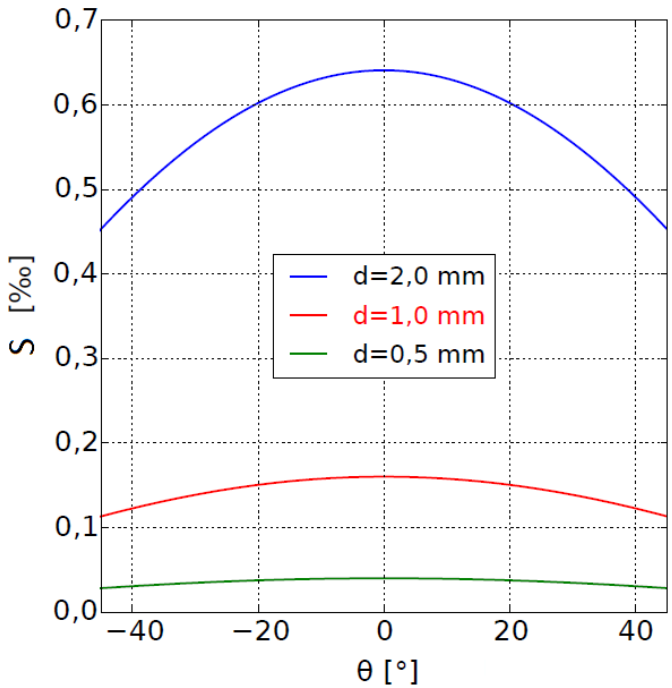
$$S = \frac{detected\ X-rays}{all\ emitted\ X-rays} \approx \frac{d_T^2 cos^3\theta}{16D^2}$$

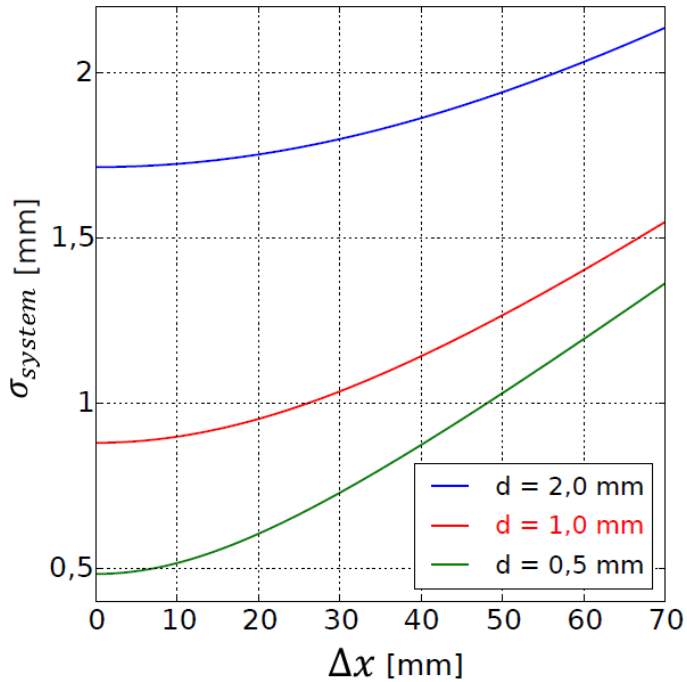
vignetting!

A.L.M. Silva et al. Spectrochimica Acta Part B 66 (2011) 308–313

R. Accorsi, S. D. Metzler IEEE TRANSACTIONS ON MEDICAL IMAGING, VOL. 23, NO. 6, JUNE 2004

Full field EDXRF imaging – pinhole optics





- Low cost
- Infinite depth of field
- Adjustable magnification

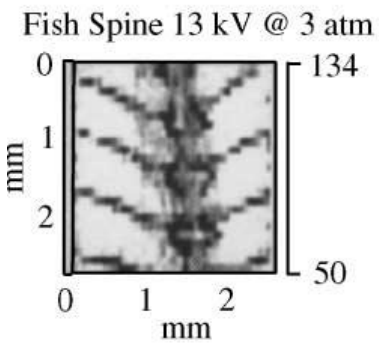
- Very low efficiency
- Vignetting

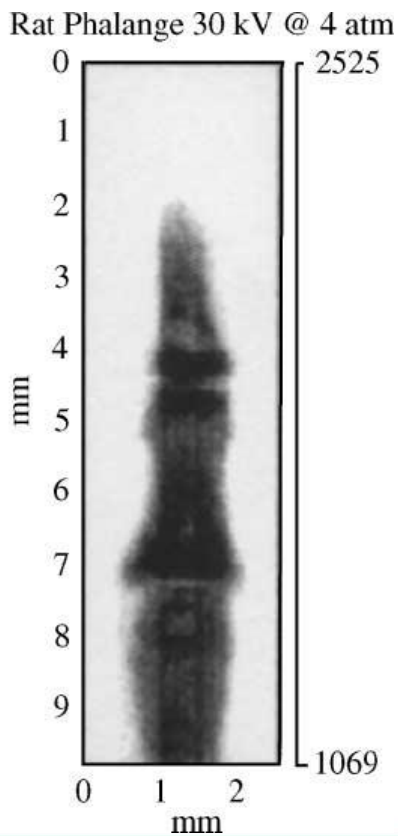
A. Zielińska, PhD thesis (in Polish), Krakow 2014

Gaseous detectors in X-ray imaging – early works

M. Li et al. / NIM A 471 (2001) 215–221

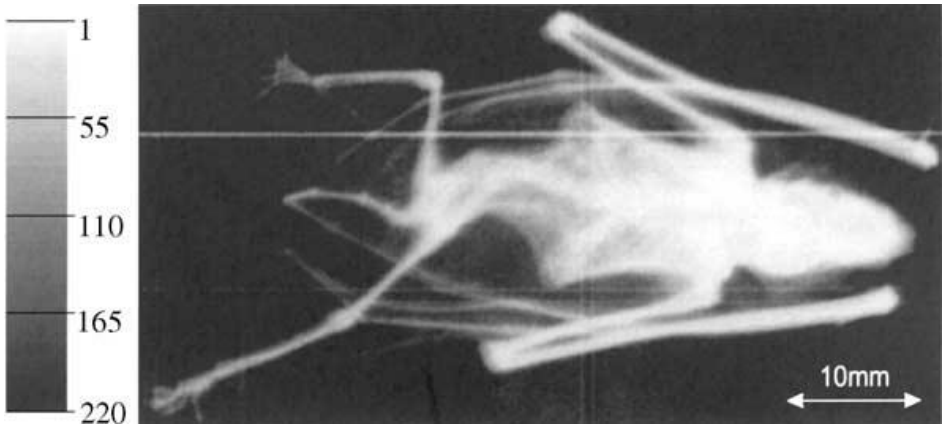
single-GEM (3.9mm Xe)
13/30 keV radiography





S. Bachmann et al. / NIMA 478 (2002) 104–108

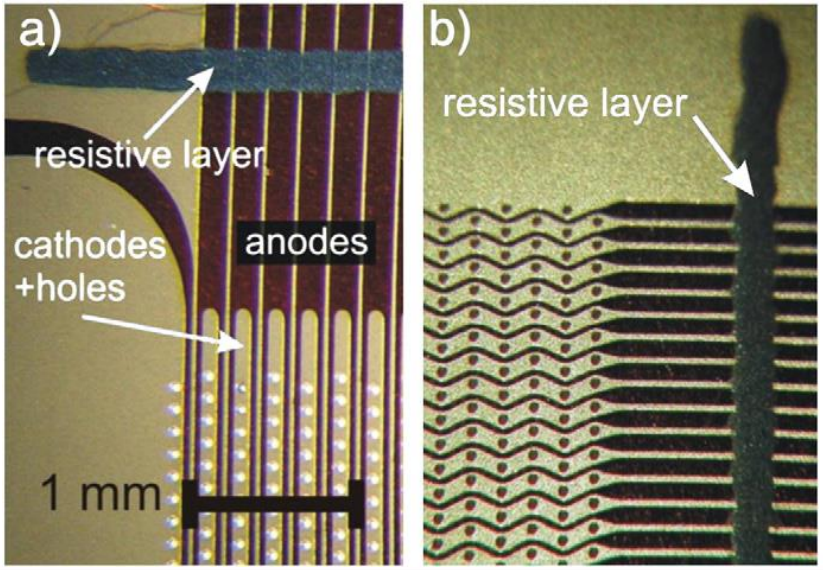
double-GEM (3mm Ar)
8keV radiography of bat



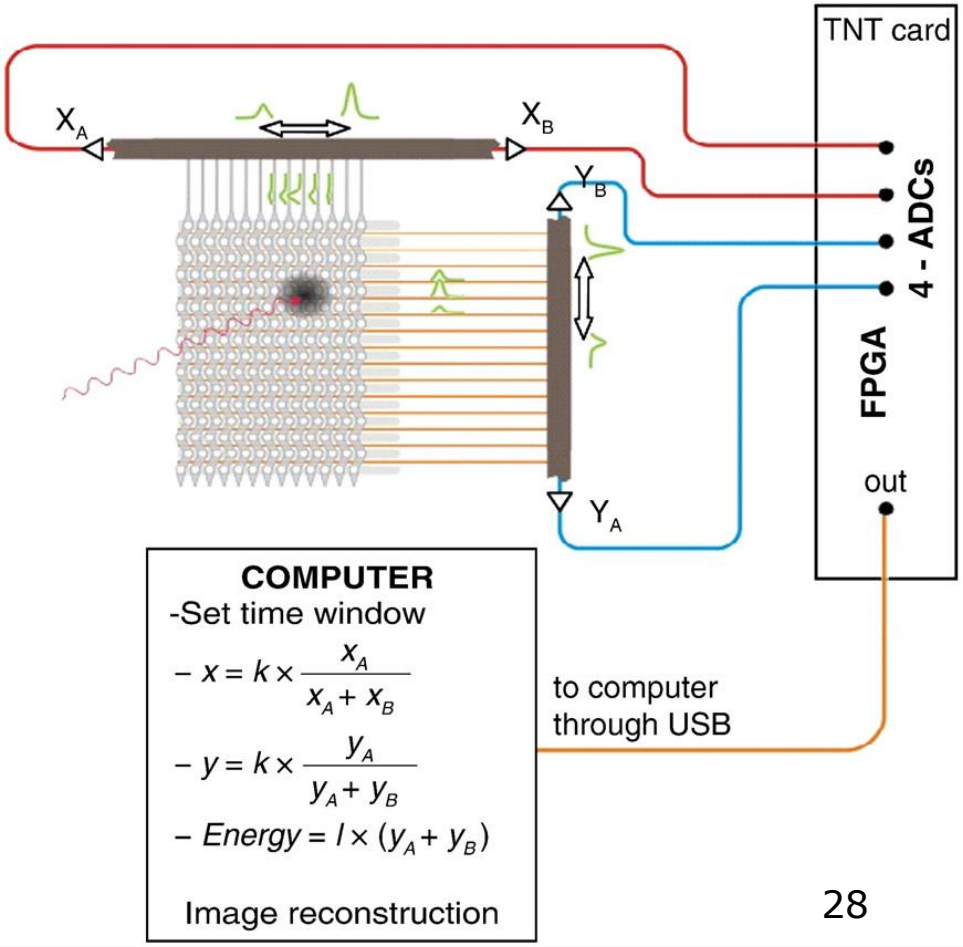
Full field EDXRF with gaseous detectors

H. Natal da Luz et al. Trans. Nucl. Sci. 55 (2008) 2341
A. L. M. Silva et al. Anal Bioanal Chem (2009) 395:2073–2080
J. F. C. A. Veloso et al. Spectrochimica Acta Part B 65 (2010) 241–247
A. L. M. Silva et al. Spectrochimica Acta Part B 66 (2011) 308–313
A. L. M. Silva et al. Spectrochimica Acta Part B 86 (2013) 115–122

- 28 x 28 mm^2 MHSP detector
- 3 mm Xe
- σ_x = 130 µm; σ_Y = 250 µm
- Counting rate up to 0.5 Mhz
- E resolution: 825 eV (5.9 keV)



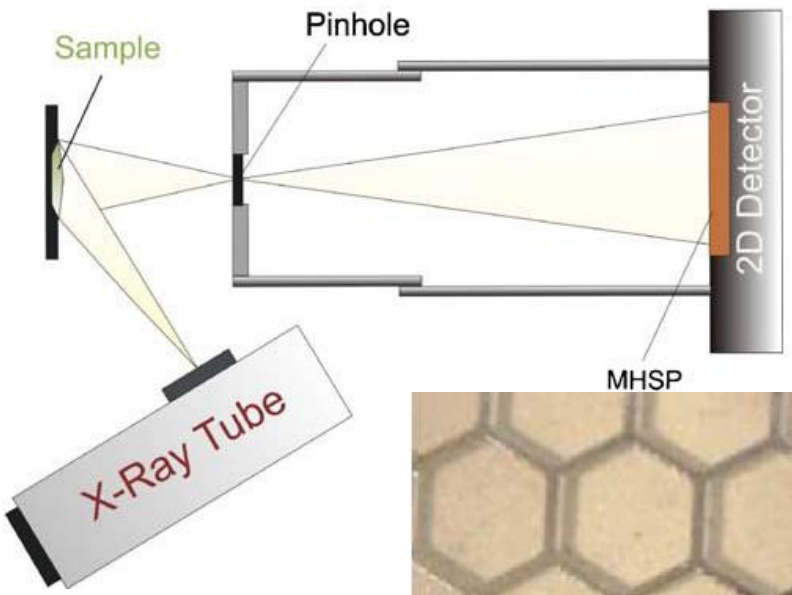
2D readout based on
resistive signal division principle

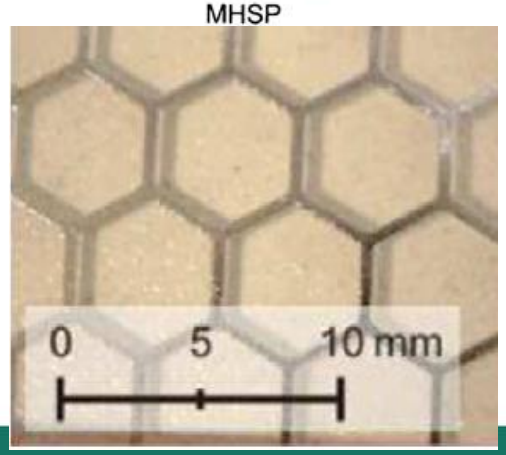


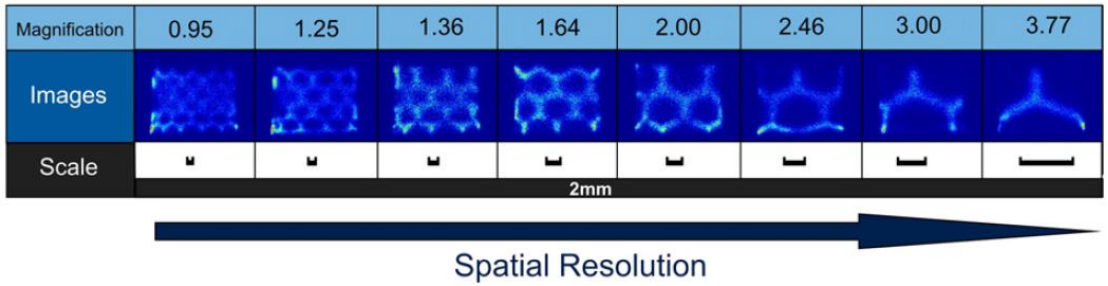
Full field EDXRF with gaseous detectors

MHSP detector experimental setup:
200 μm tungsten pinhole with telescopic tube
50 W molybdenum X-ray tube

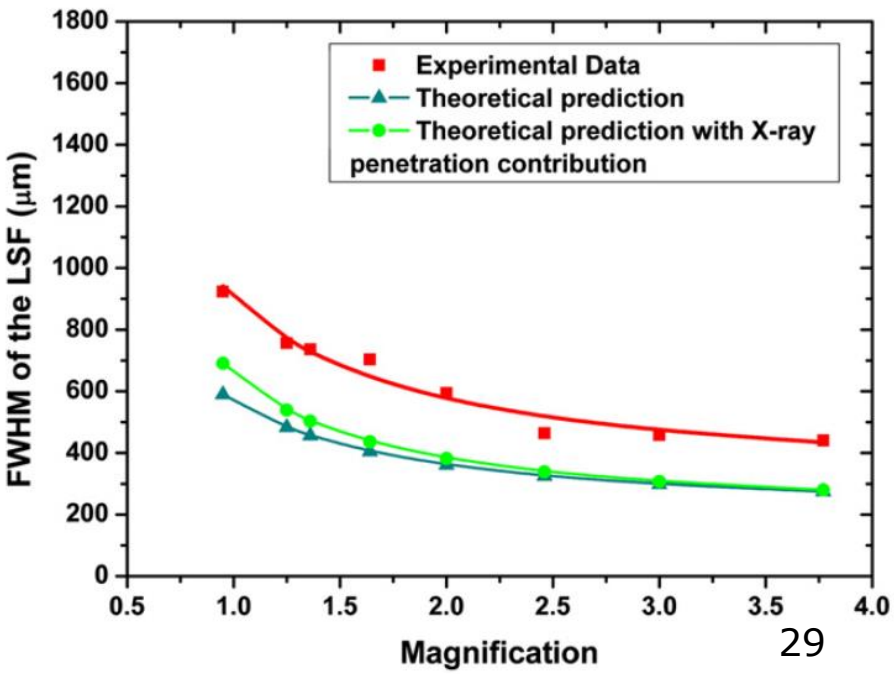
A. L. M. Silva et al. Spectrochimica Acta Part B 66 (2011) 308–313







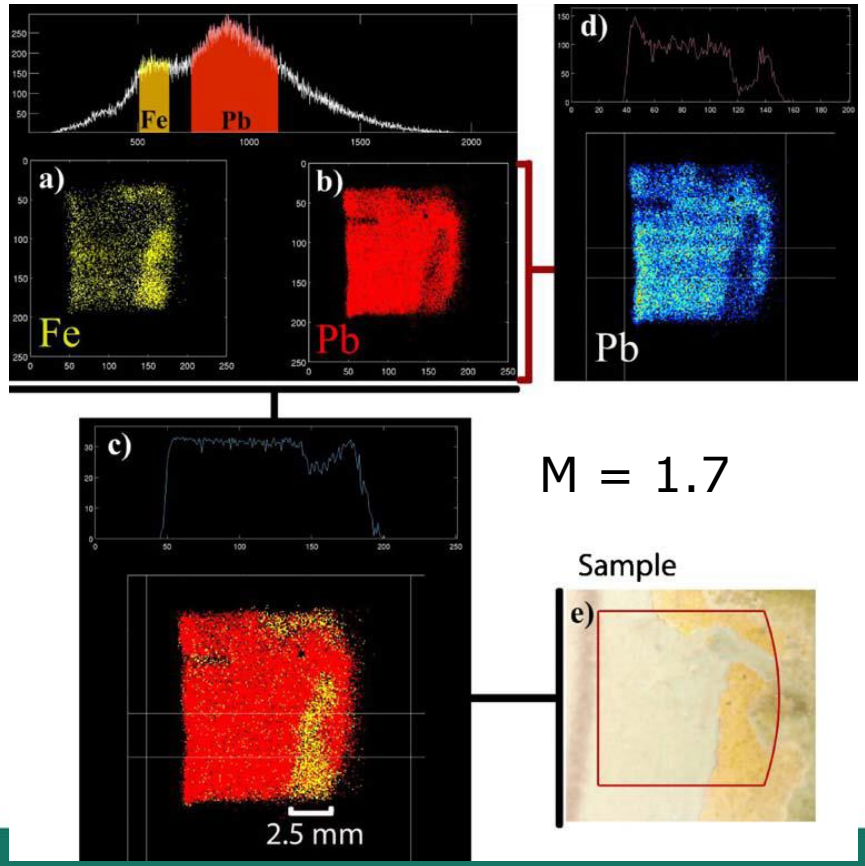
Spatial Resolution



Full field EDXRF with gaseous detectors

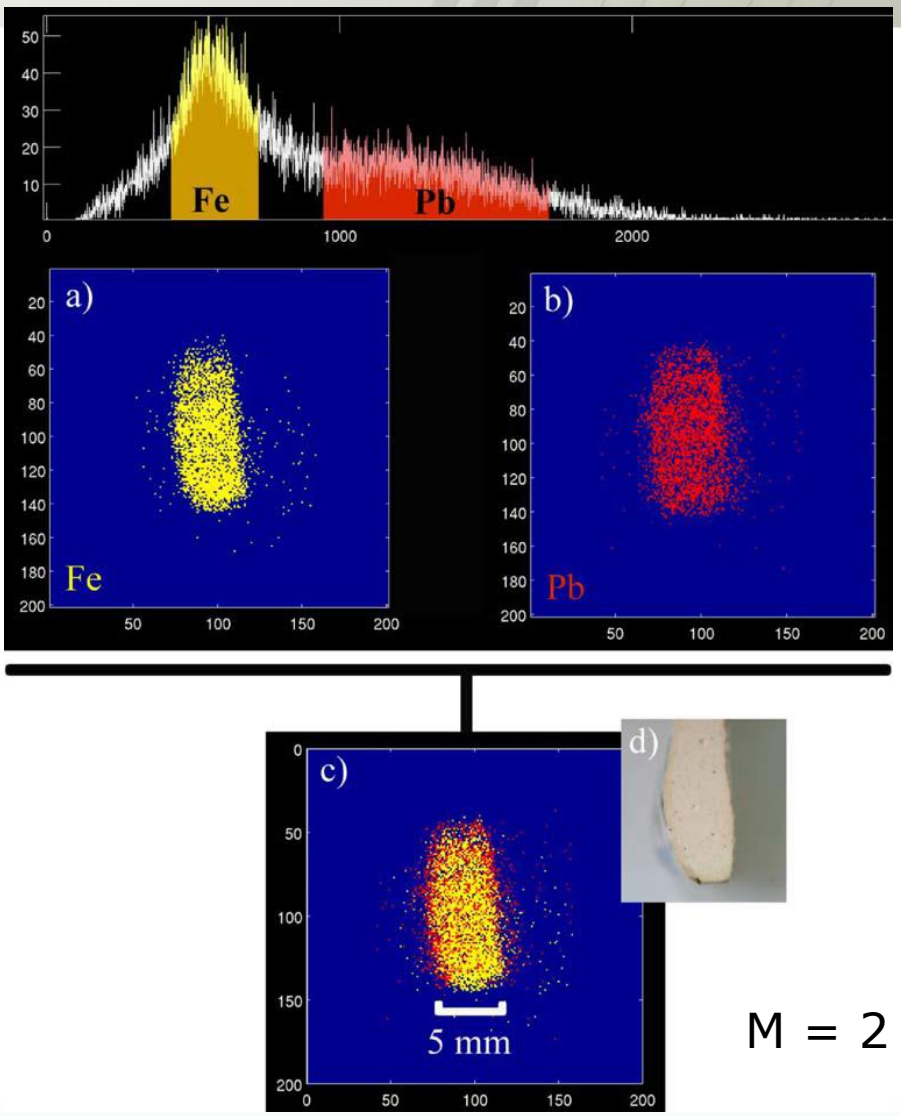
A. L. M. Silva et al. Anal Bioanal Chem (2009) 395:2073–2080

Cross sections of 18th century faiences from Coimbra (Portugal)
20kV / 1mA



M = 1.7

Sample

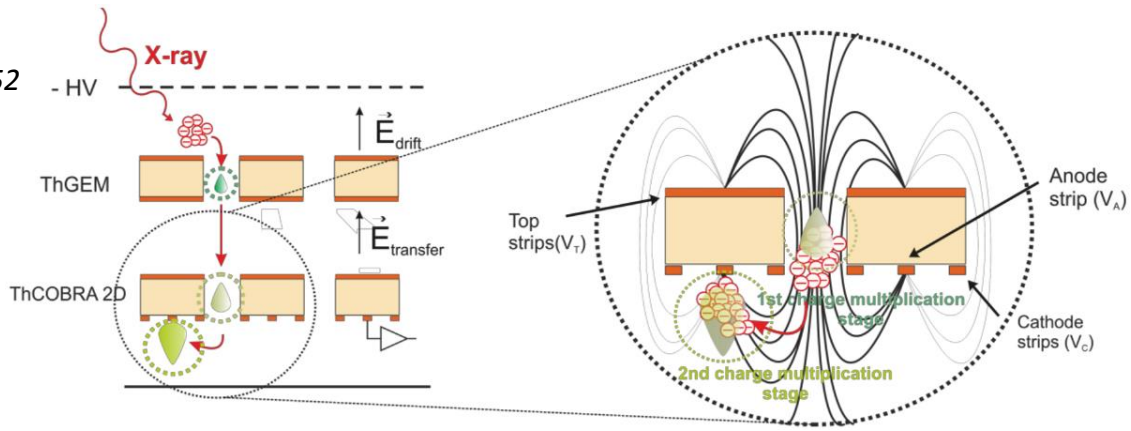


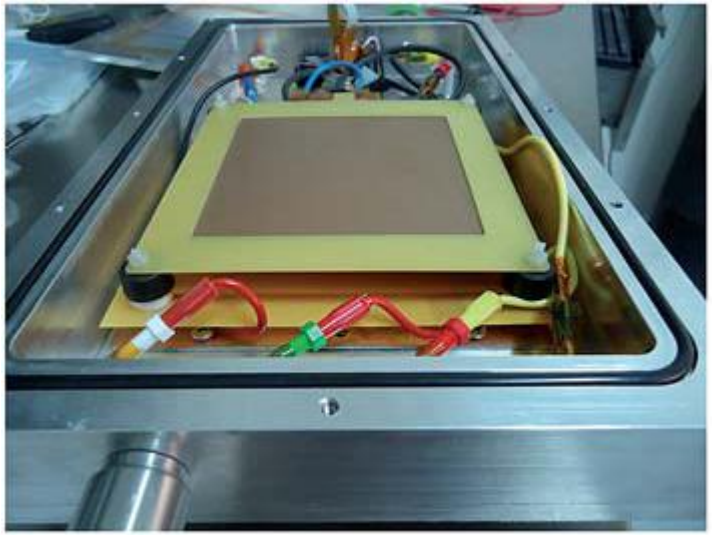
M = 2

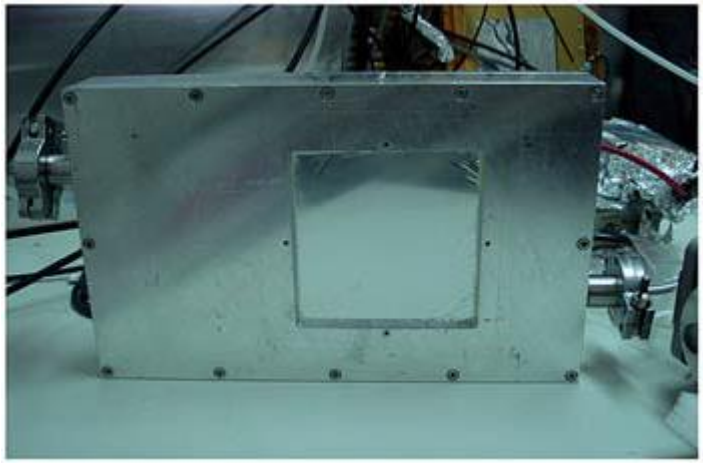
Full field EDXRF with gaseous detectors

A. L. M. Silva et al. 2013 JINST 8 P05016
L. F. N. D Carramate et al. 2015 JINST 10 P01003
A. L. M. Silva et al. J. Anal. At. Spectrom., 2015, 30, 343–352

100x100 mm^2
ThGEM + ThCOBRA
Ne/CH_4





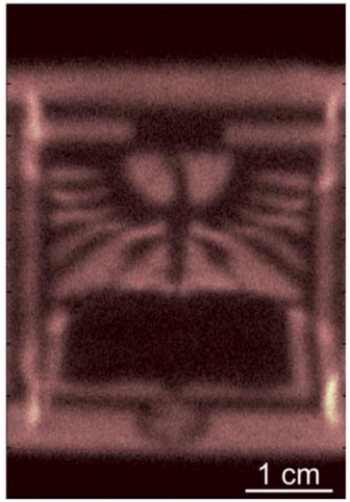


Full field EDXRF with gaseous detectors

A. L. M. Silva et al. 2013 JINST 8 P05016

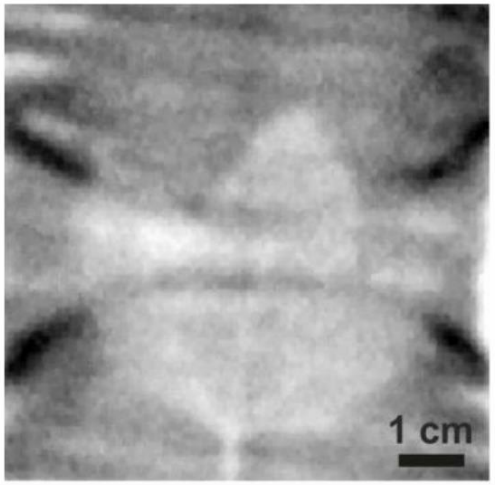
100x100 mm^2
ThGEM + ThCOBRA
20.3% resolution for Cu-Kα
Detector intrinsic resolution:
σ_x = 0.97 mm; σ_Y = 1.16 mm

50 W Mo X-ray tube



20 kV, 320 mA, 20 min

5.8 kV, 630 mA, 20 min



Full field EDXRF with gaseous detectors

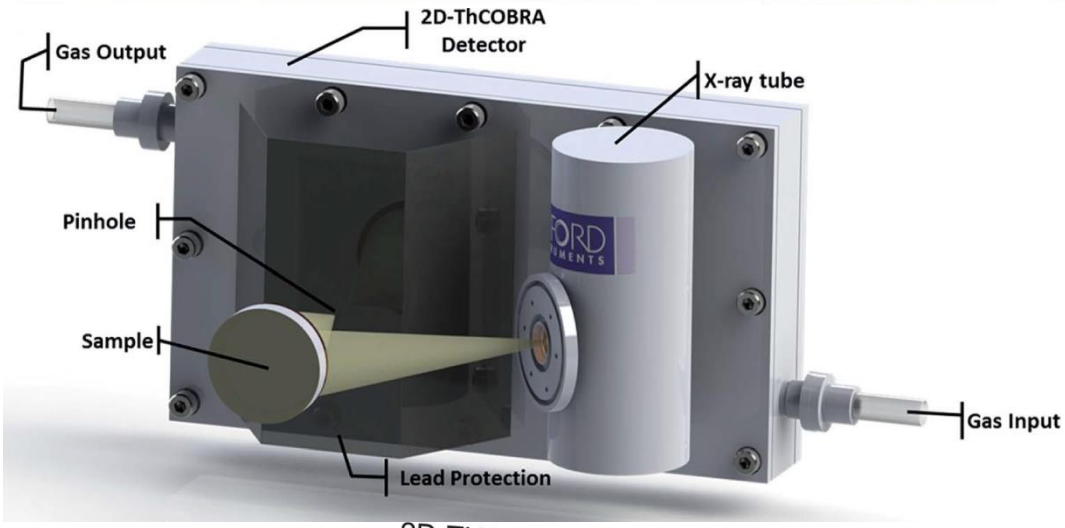
A. L. M. Silva et al. J. Anal. At. Spectrom., 2015, 30, 343–352

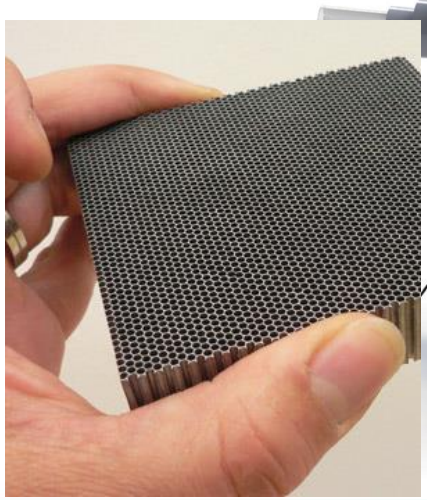
100x100 mm^2
ThGEM + ThCOBRA

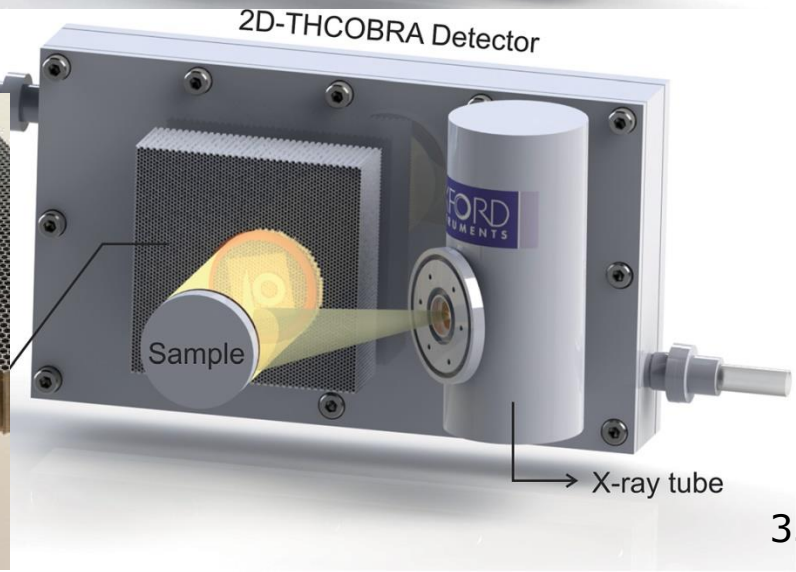
OXFORD 50 W X-ray tube

Pinhole optics:
0.6 – 2 mm, 300µm lead foil

Multihole lead collimator:
honeycomb structure
2mm hole, 4cm thickness,
0.2 mm wall thickness

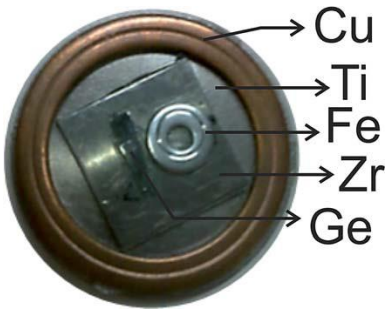




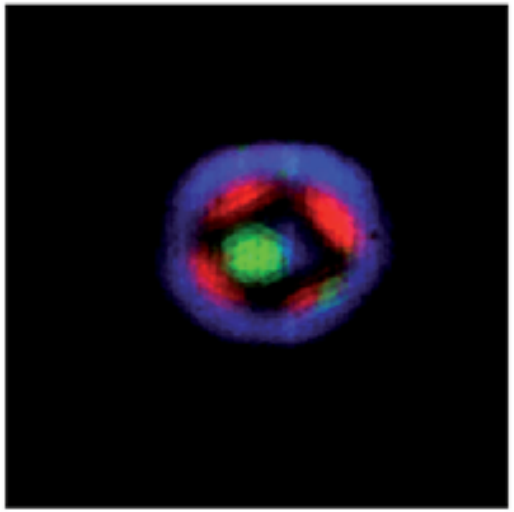


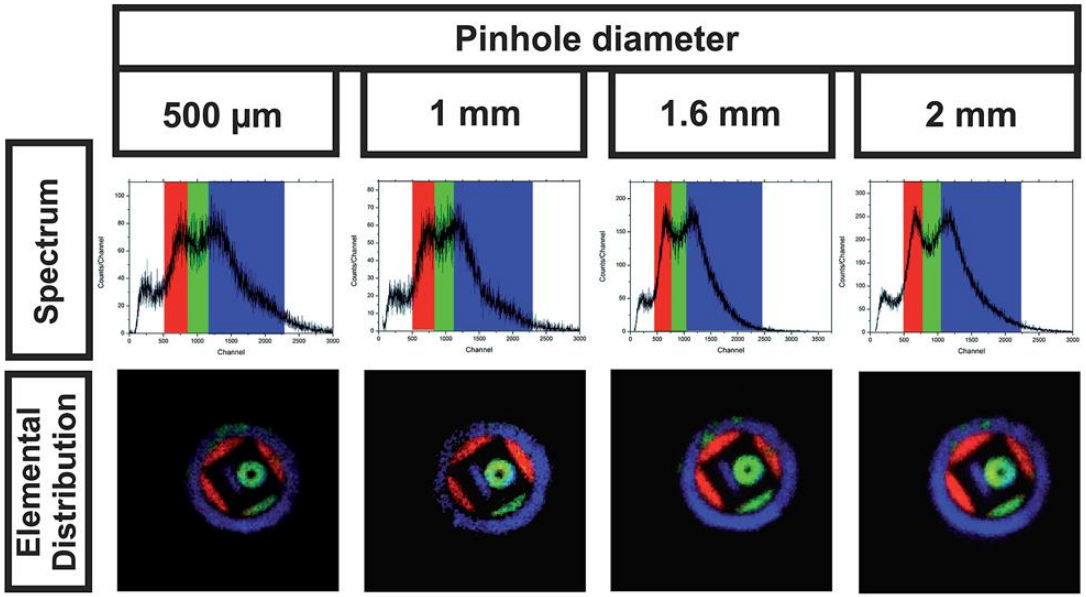
Full field EDXRF with gaseous detectors

A. L. M. Silva et al. J. Anal. At. Spectrom., 2015, 30, 343–352



Multihole collimator result:
40x more intensity than 1mm pinhole
poor resolution, honeycomb artefacts





15 min exposure, M = 1, 20 kV, 1mA

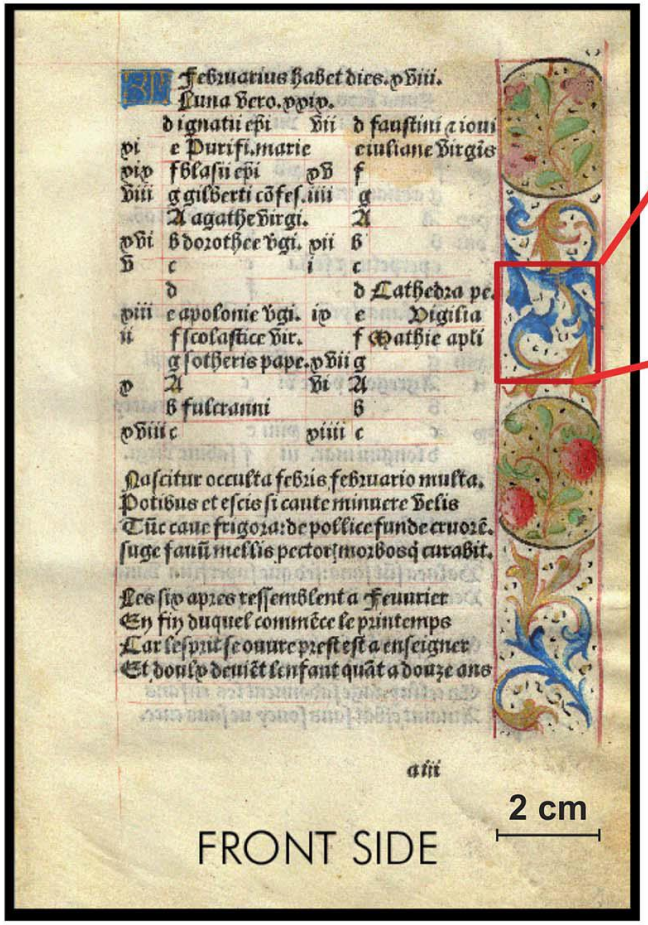
Pinhole optics result:
500 µm resolution at M=3 and 0.5 mm pinhole

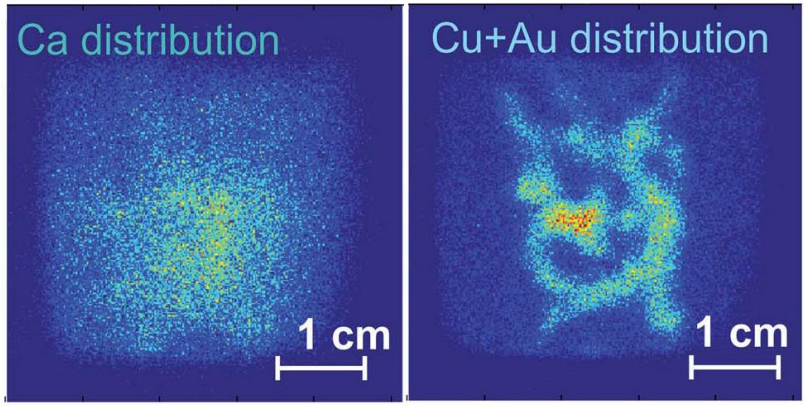
Cu+Ge, Ti, Fe

Full field EDXRF with gaseous detectors

A. L. M. Silva et al. J. Anal. At. Spectrom., 2015, 30, 343–352

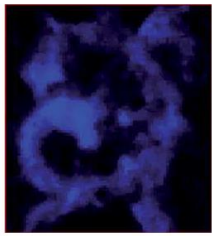
ornaments from illuminated 15th–16th century Book of Tides

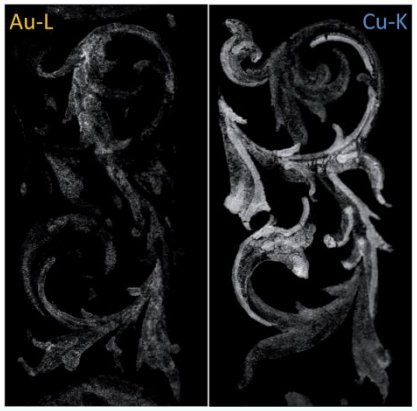




low power tube, 25kV, 1 mA, 15 min, M = 2.5

high power tube,
30kV, 50 mA, M = 2.7



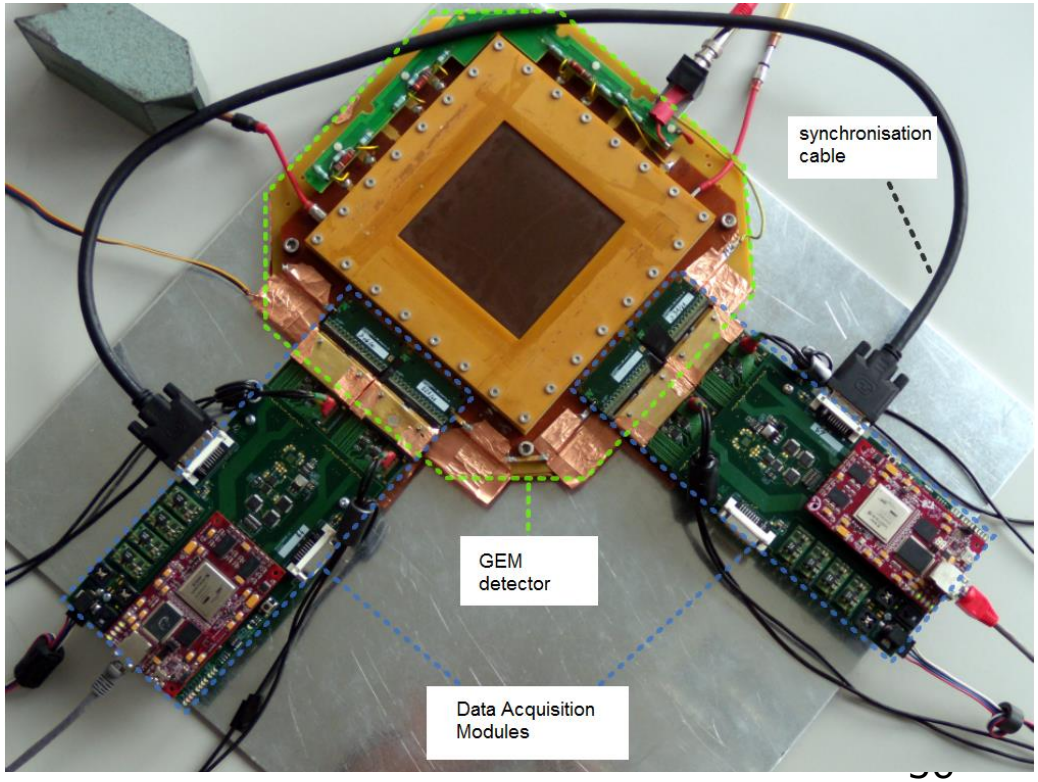


Full field EDXRF with gaseous detectors

A. Zielinska et al. 2013 JINST 8 P10011
A. Zielinska, PhD thesis (in Polish), Krakow 2014

- 100x100 mm^2 TGEM
- 3 mm Ar/CO_2
- 2D readout by crossed electrodes (0.4 pitch – effective pitch: 0.8 mm)
- 32-channel Application Specific Integrated Circuits – 4 per coordinate
- (2 x 128 readout channels)
- DAQ modules: ADC + FPGA

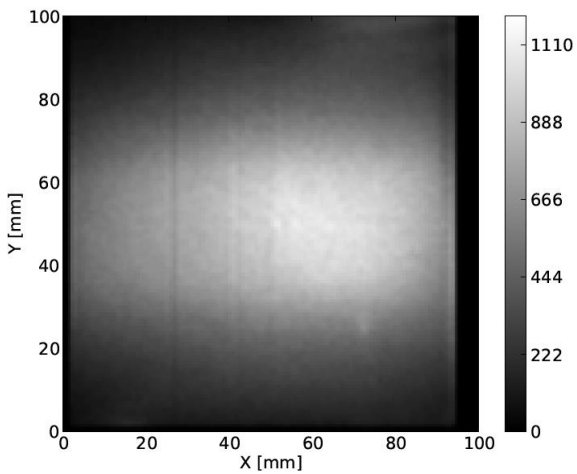
- Counting rate up to 5MHz
- Energy resolution: 20% (5.9 keV)
- σ = 230 µm

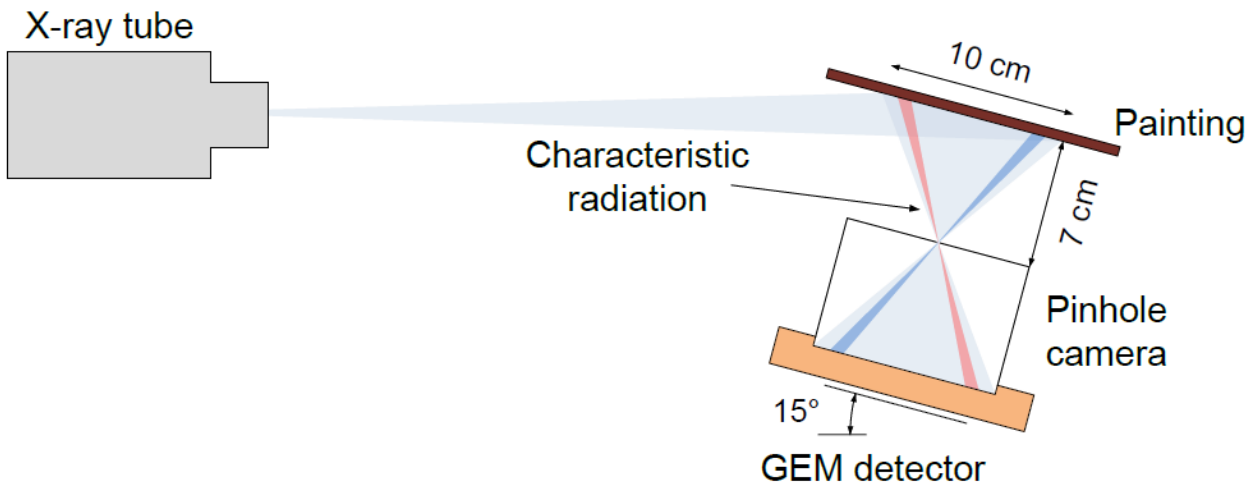


Full field EDXRF with gaseous detectors

A. Zielinska et al. 2013 JINST 8 P10011
A. Zielinska, PhD thesis (in Polish), Krakow 2014

TGEM detector experimental setup:
High power Mo X-ray tube – beam size 3x10 cm^2
30 kV / 40 mA
Source – sample ~ 90cm
0.5 - 2 mm Cd pinhole
Fixed M = 1





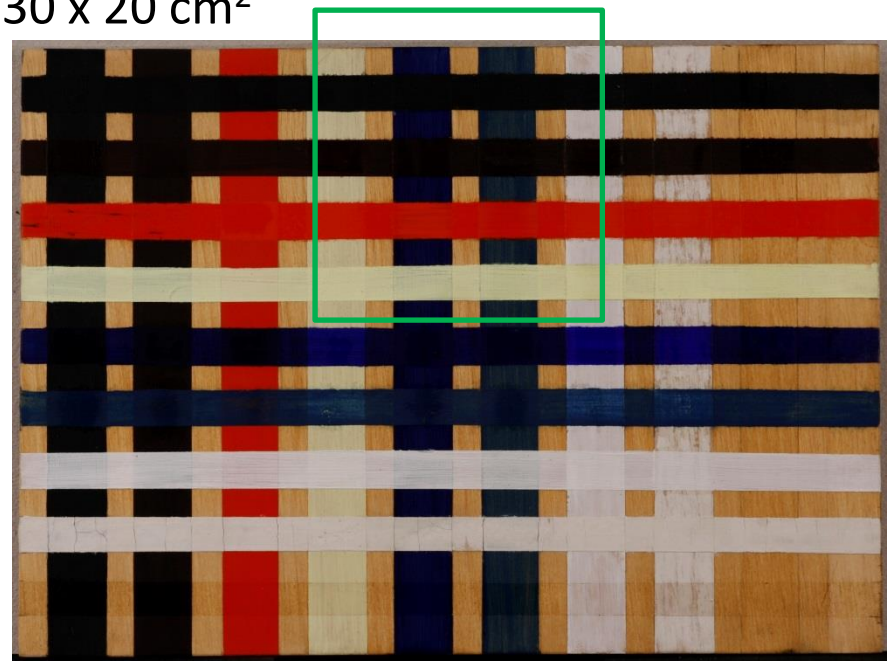
Full field EDXRF with gaseous detectors

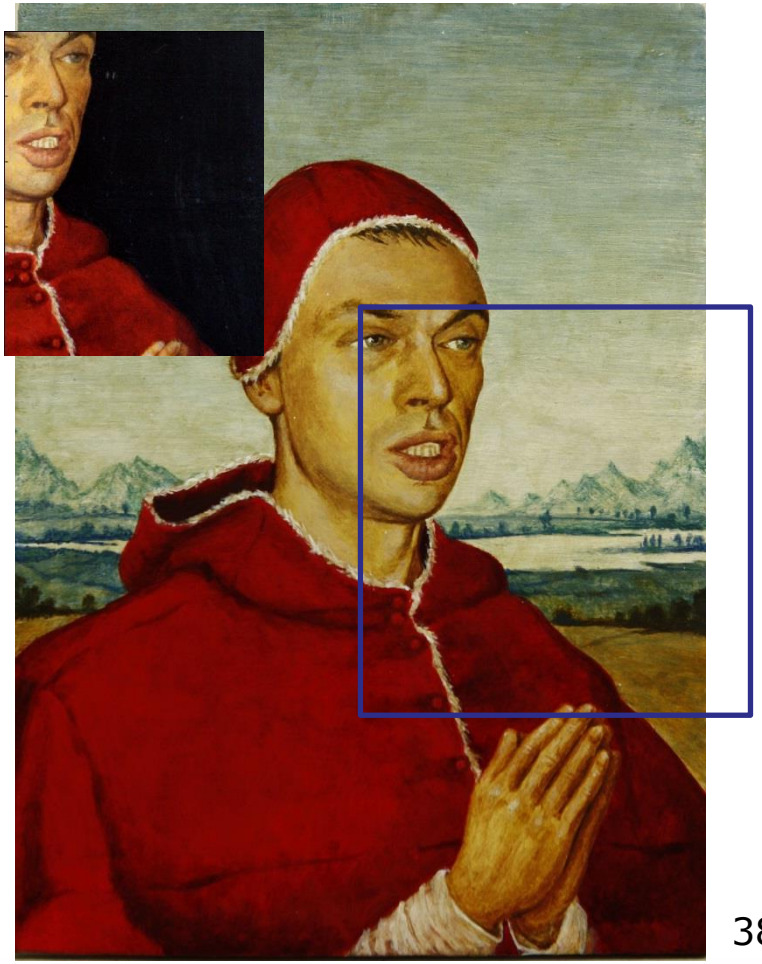
Test samples on wooden panels:

- Stripe pattern with different pigments
- Mock-up painting „Man in a red coat”

A. Zielinska et al. 2013 JINST 8 P10011
A. Zielinska, PhD thesis (in Polish), Krakow 2014

30 x 20 cm^2



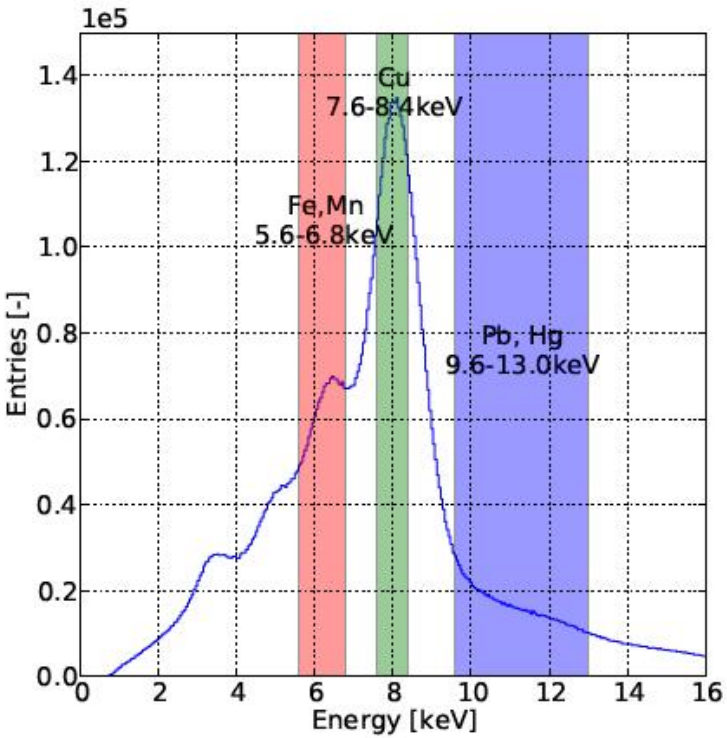


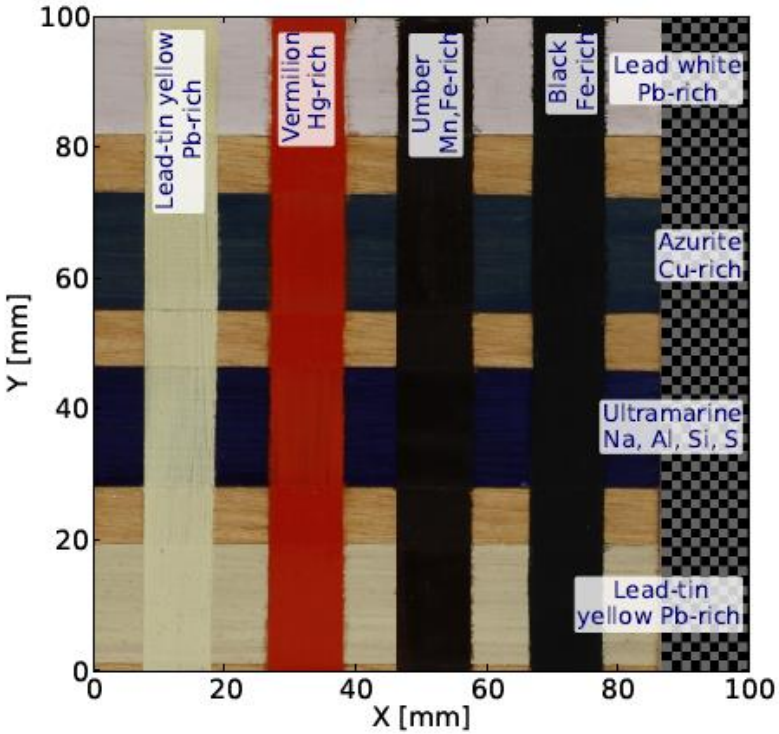
22 x 16 cm^2

Full field EDXRF with gaseous detectors

Results: stripe pattern 50 min exposure

A. Zielinska et al. 2013 JINST 8 P10011
A. Zielinska, PhD thesis (in Polish), Krakow 2014



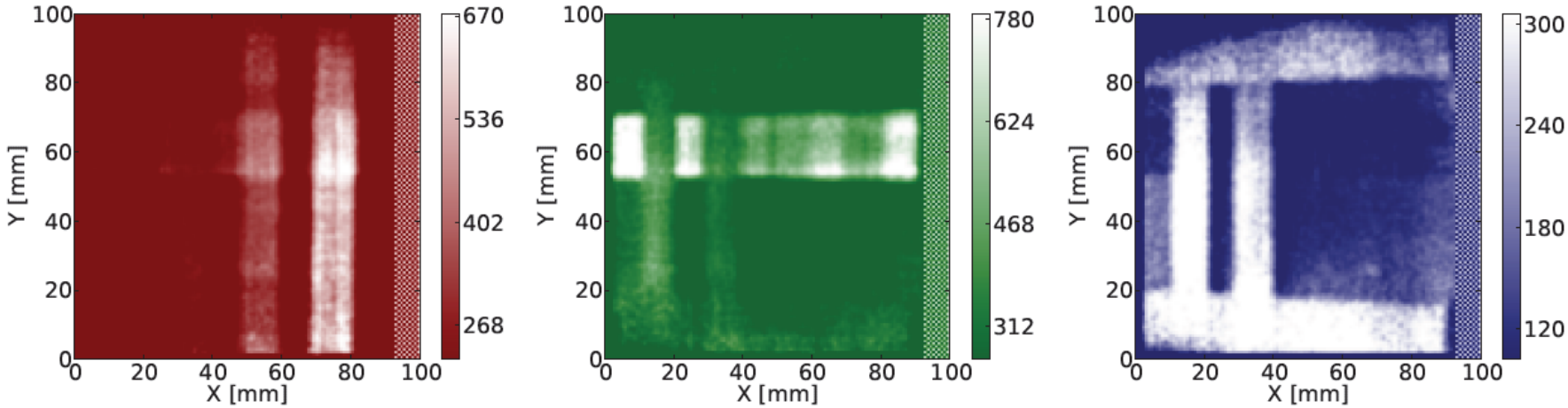


Full field EDXRF with gaseous detectors

Results: stripe pattern 50 min exposure

A. Zielinska et al. 2013 JINST 8 P10011
A. Zielinska, PhD thesis (in Polish), Krakow 2014

average registered rate: 3.2 kHz



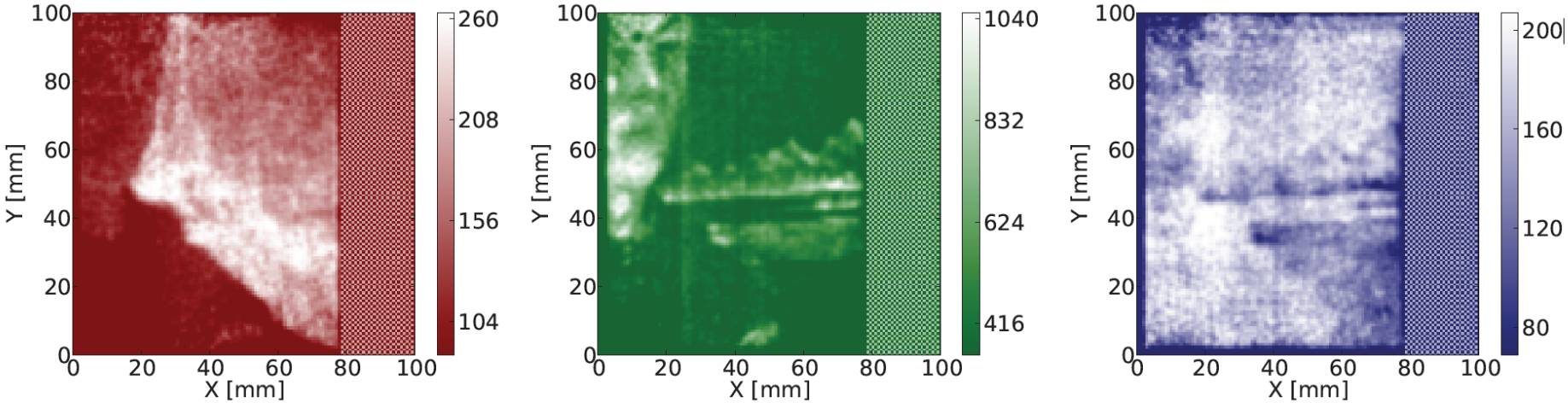
(a) Fe and Mn map (5.5-6.8 keV). **(b)** Cu map (7.6-8.4 keV). **(c)** Pb and Hg map (9.6-13 keV).

Full field EDXRF with gaseous detectors

Results: mock-up painting 5.5 h exposure

A. Zielinska et al. 2013 JINST 8 P10011
A. Zielinska, PhD thesis (in Polish), Krakow 2014

average registered rate: 450 Hz



(a) Fe map (6.0-6.8 keV). **(b)** Cu and Zn map (7.6-9.0 keV). **(c)** Pb map (10.0-13.0 keV).

Full field EDXRF with gaseous detectors

radiography
W X-ray tube
20kV 40mA
10 min
23.8 kHz

A. Zielinska, PhD thesis (in Polish), Krakow 2014

Goals and perspectives:

- use of Xe
- replacement of entrance eletrode (5 µm Cu)
  - problem with excitation of copper
- developement of dedicated readout electronics
- optimisation of exctitation system
- creation of portable device and scanner

Conclusions

	BI-DD-CCD	pnCCD	gaseous
Thickness	40 µm Si	450 µm Si	1-5 mm Ne/Ar/Xe
QE	90% (5 keV) 8.5 % (15 keV)	100% (5 keV) 64 % (15 keV)	86% (5 keV, Xe 5 mm) 15 % (15 keV, Xe 5 mm)
Sensor size	13x13 mm^2	12.7x12.7 mm^2	**100x100 mm^2**
Energy resolution	133 eV (at Mn-Kα)	152 eV (at Mn-Kα)	1180 eV (at Mn-Kα)
Pixel size	13 µm	48 µm	(0.4 mm)
Max count rate	<10kcps	600 kcps	**5 Mcps**
System resolution (at 1:1 magnification)	~150 µm	50 µm	<500 µm (250 µm achievable)
Optics	Pinhole	Polycapillary	Pinhole
Cost	??	~300 000 €	**~3000 € (TGEM)** **~300 € (ThCOBRA)**

Acknowledgements

AGH – UST
W. Dabrowski, M. Lankosz, A. Zielinska, B. Mindur, P. Wiacek,
K. Swietek

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L. Bratasz, B. Swiatkowska, P. Fraczek

National Centre for Research and Development (NCBR)
Applied Research Programme

Thank you for your attention!