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

\[ N = \frac{E}{W}, W \approx 25 \text{ eV/pair} \]

\[ N \approx 200 \text{ (for 5 keV)} \]

3) Ions and electrons drifts towards electrodes in electric field
Drift of charge induce current:

\[ i_k(t) = Nq v_k(t) \cdot \Psi(\vec{r}) \quad v_k(t) = \mu_k E(\vec{r}(t)) \]

mobility:

\[ \mu_{\text{electron}} \approx \text{cm/\mu s} \]
\[ \mu_{\text{ion}} \approx \text{cm/ms} \]

For flat ionisation chamber:

Charge collection time:

\[ T_{\text{ion}} \gg T_{\text{electron}} \]

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 ($\sim 10^6 \text{ V/m}$) 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

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
- Deexcitation of molecules – creation of UV photons
- Absorption of UV photons – creation of new ion pairs
- New avalanche 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 requires 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 length : 20 mm
- Spatial resolution limit : 300 μm
  (pitch / \sqrt{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)

\[ \frac{y}{L} = \frac{Q_B}{Q_B + Q_A} \]

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

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

second coordinate – segmenting of bottom side of the insulator

Micro Strip Gas Chamber

- Large area 100 X 100 mm²
- Small distance between strips – spatial resolution ~30 µm
- Short path to travel for ions – short pulses – high speed (10⁶ Hz/mm²)
- Very high field – sparks (aging or damage)
- Charging of substrate material – gain fluctuations

https://gdd.web.cern.ch/GDD/msgc.html
I. Giomatri - 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.
F. Sauli – 1997

- Cu electrodes separated with kapton (litography + etching)
- 100 holes / mm²
- 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 ($10^5$)
- worse spatial resolution

F. Sauli, Nucl. Instr. and Meth. A386(1997)531
Single GEM gives small gain (up to $10^2$) – solution: use several GEM foils

- ion movement negligible – very fast electron signal (<40 ns)
- count rate up to $10^6$ Hz/mm$^2$
- resolution < 80 µm
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
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 mm²
1242x1152px
res: ~400 eV

hybrid-pixel
14x14 mm²
256x256px
res: 3 – 5 keV

Photo of the sample in the optical range
Resulting spectral-selective image of the sample surface

pure metallic Ti/Cu/Ge sample

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 mm² 1024x1024 px
FOV: 2.5x2.5mm² - 4x4 cm²

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


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

Full field EDXRF imaging – pinhole optics

Magnification: \[ M = \frac{d}{D} \]

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

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

\[ \sigma_{\text{det}} = \frac{p}{\sqrt{12}} \]

\[ \sigma_{\Delta x} = \frac{d'\Delta x/d_{T}}{\sqrt{8\ln(2)M}} \left( 1 + \frac{1}{M} \right) \]

Sensitivity:

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

vignetting!

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
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


double-GEM (3mm Ar)
8keV radiography of bat
Full field EDXRF with gaseous detectors


- 28 x 28 mm² MHSP detector
- 3 mm Xe
- σₓ = 130 µm; σᵧ = 250 µm
- Counting rate up to 0.5 Mhz
- E resolution: 825 eV (5.9 keV)

2D readout based on resistive signal division principle

![Diagram of EDXRF setup with MHSP detector and resistive layer connections.](image)
Full field EDXRF with gaseous detectors

MHSP detector experimental setup:
200 µm tungsten pinhole with telescopic tube
50 W molybdenum X-ray tube
Full field EDXRF with gaseous detectors


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

M = 1.7

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

100x100 mm²
ThGEM + ThCOBRA
Ne/CH₄
Full field EDXRF with gaseous detectors

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

100x100 mm²
ThGEM + ThCOBRA
20.3% resolution for Cu-Kα
Detector intrinsic resolution:
\( \sigma_x = 0.97 \text{ mm}; \sigma_y = 1.16 \text{ mm} \)

50 W Mo X-ray tube

5.8 kV, 630 mA, 20 min

20 kV, 320 mA, 20 min
Full field EDXRF with gaseous detectors

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


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

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

Cu+Ge, Ti, Fe
Full field EDXRF with gaseous detectors


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

- 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)
- $\sigma = 230 \mu$m
Full field EDXRF with gaseous detectors

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

A. Zielinska et al. 2013 JINST 8 P10011
A. Zielinska, PhD thesis (in Polish), Krakow 2014
Full field EDXRF with gaseous detectors

Test samples on wooden panels:
• Stripe pattern with different pigments
• Mock-up painting „Man in a red coat”

30 x 20 cm²

22 x 16 cm²
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

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).
Results: mock-up painting 5.5 h exposure

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

Goals and perspectives:

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

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

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

<table>
<thead>
<tr>
<th></th>
<th>BI-DD-CCD</th>
<th>pnCCD</th>
<th>gaseous</th>
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</thead>
<tbody>
<tr>
<td>Thickness</td>
<td>40 µm Si</td>
<td>450 µm Si</td>
<td>1-5 mm Ne/Ar/Xe</td>
</tr>
<tr>
<td>QE</td>
<td>90% (5 keV) 8.5% (15 keV)</td>
<td>100% (5 keV) 64% (15 keV)</td>
<td>86% (5 keV, Xe 5 mm) 15% (15 keV, Xe 5 mm)</td>
</tr>
<tr>
<td>Sensor size</td>
<td>13x13 mm²</td>
<td>12.7x12.7 mm²</td>
<td>100x100 mm²</td>
</tr>
<tr>
<td>Energy resolution</td>
<td>133 eV (at Mn-Kα)</td>
<td>152 eV (at Mn-Kα)</td>
<td>1180 eV (at Mn-Kα)</td>
</tr>
<tr>
<td>Pixel size</td>
<td>13 µm</td>
<td>48 µm</td>
<td>(0.4 mm)</td>
</tr>
<tr>
<td>Max count rate</td>
<td>&lt;10kcps</td>
<td>600 kcps</td>
<td>5 Mcps</td>
</tr>
<tr>
<td>System resolution</td>
<td>~150 µm</td>
<td>50 µm</td>
<td>&lt;500 µm (250 µm achievable)</td>
</tr>
<tr>
<td>(at 1:1 magnification)</td>
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<tr>
<td>Optics</td>
<td>Pinhole</td>
<td>Polycapillary</td>
<td>Pinhole</td>
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<tr>
<td>Cost</td>
<td>??</td>
<td>~300 000 €</td>
<td>~3000 € (TGEM) ~300 € (ThCOBRA)</td>
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Thank you for your attention!