Development of research around 1.7 MV Pelletron in Jyväskylä during five years of operation

Mikko Laitinen, J. Julin, L. Mättö, M. Napari, N. Puttaraksa, T. Sajavaara

Department of Physics, P.O.B 35, FIN-40014 University of Jyväskylä, Finland
email: mikko.i.laitinen@jyu.fi
Outlook

- Background: Who we are
- Moving the 1.7MV Pelletron to Jyväskylä
- Water leak at the SF6 heat exchanger
- From corona needles to resistors
- Turbopumped stripper system installation
- Beamlines and recent developments
- ‘Contaminants’ after analyzing magnet
University of Jyväskylä, Department of Physics (JYFL)

- **University**: Staff 2600, 15000 Students, 217 M€ turnover
- **Department**: Personnel 190, including 85 PhD students

Physics department main research areas:
- Nuclear and accelerator based physics
- One team out of 8: Accelerator based materials physics
- Materials physics
- High-energy physics

**NordForsk 2011**: Comparing Research at Nordic Universities using Bibliometric Indicators

Among 30 Nordic Universities,

*JyU is among the top four universities in Physics + Mathematics*

(“second place” after Aarhus U.)
Accelerator based materials physics

- 1 Senior, no other staff, including engineers
- 4-5 PhD students (for example: ion beam lithography, detector development, direct signal digitization by fast digitizers)
- 4-6 Master (and bachelor) thesis students

24/7 working diffusion cloud chamber for physics department
Permanent exhibition: Masters thesis project

Commercial system price tag: 25-50k€ depending on size
MOVING THE 1.7MV PELLETRON TO JYVÄSKYLÄ
Acquisition of the Pelletron accelerator

- A coffee table rumor was heard in late 2006… … and quickly confirmed by indirect route

- The technical research center of Finland (VTT) had little usage of their accelerator and needed the room space for cleanroom extension. … And our group in Jyväskylä needed an accelerator.

- 1.7MV 5SDH-2 Pelletron (serial number 002, made in 1985) with one (Alphatross) ion source and one beam line was donated to JYU by VTT
Moving the Accelerator from VTT

- SF6 purity 88%!
- No moisture 19ppm
- Purity analysis by Solvay Germany

<table>
<thead>
<tr>
<th>Herkunft</th>
<th>Gebinde</th>
<th>Gewicht (kg)</th>
<th>Anteil SF6</th>
<th>SF6</th>
<th>Luft</th>
<th>CF4</th>
<th>CO2</th>
<th>SO2F2</th>
<th>SO2</th>
<th>S2F10</th>
<th>SF5-O-SF5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wikeström, Helsinki</td>
<td>Flasche 72356</td>
<td>28</td>
<td>18</td>
<td>65.2939</td>
<td>34.706</td>
<td>0.0206</td>
<td>0.00009</td>
<td>0.00516</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wikeström, Helsinki</td>
<td>Flasche 74390</td>
<td>21</td>
<td>16</td>
<td>75.2364</td>
<td>24.231</td>
<td>0.0699</td>
<td>0.0226</td>
<td>0.28837</td>
<td>0.01014</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wikeström, Helsinki</td>
<td>Flasche 12780</td>
<td>30</td>
<td>28</td>
<td>93.8826</td>
<td>6.1131</td>
<td>0.001</td>
<td>0.00009</td>
<td>0.00516</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Wikeström, Helsinki</td>
<td>Flasche 9977952</td>
<td>33</td>
<td>30</td>
<td>89.6052</td>
<td>10.395</td>
<td>0.0022</td>
<td>0.00009</td>
<td>0.00516</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

S2F10 was considered a potential chemical warfare agent in World War II because it does not produce lacrimation or skin irritation, thus providing little warning of exposure. LD50 levels about 15-25 ppm

Monday 18th of September 2006
Moving the Accelerator from VTT

Tuesday 19th of September 2006
Arriving at Jyväskylä after 330km

25th of September 2006
Building of the Pelletron lab

Target hall
30th-31st November 2006
Installing of the Pelletron

14th of December 2006
Installing of the Pelletron

17th of December 2006
2nd of January 2007
Cleaning of the Pelletron

18th of January 2007
Powering up of the Pelletron acc.

6th of February 2006
Ion source cleaning + commissioning
First RBS measurements

Thin (170 nm) hydroxyapatite

- Ca/P ratio 1.18

- Beam on target: 21.2.2007
- Total commissioning time from ‘amateurs’: ~2months

Thick (550 nm) hydroxyapatite

- Ca/P ratio 1.33

- Energy (ch)
- Yield

Experimental
SIMNRA simulation
WATER LEAK AT THE SF6 HEAT EXCHANGER
Water leakage from the heat EX

- First symptom: Voltage(GVM) didn’t go DOWN when chains were turned off
- Real signal: Voltage rised very poorly, even after long “conditioning” (about 1.5 weeks from first symptoms)

11.9.2007

STARTING THE ion SOURCE AND ACCELERATOR AGAIN.

STRANGE OBSERVATION: IT USED TO BE SO THAT ONCE CHAIN
Water leakage from the heat EX

- Measuring the purity of SF6: 99.6% (27.9.2007)
- Water content in SF6: **7500 ppm**

5th of October 2007
Water leakage from the heat EX

5th of October 2007, almost a month from first symptoms
Water leakage from the heat EX

Problem verified

Water condensation @ ~7500 ppm \( \text{H}_2\text{O} \) in \( \text{SF}_6 \) @ 2.7 bar, 24°C

A lot of water out from alumina

Pumping few days of the external heat exchanger tubing with dry scroll pump.

New alumina in -> gas circulation
Conclusions and aftermath of the water leakage after tank opening + cleaning:

- There had been few cm of liquid water inside the tank bottom ("high water" -mark)
- New "pure" ion exchanged water at new lab might enhanced corrosion of the rusted Cu pipes of the original heat exchanger
- Only aftermath was most likely the GVM bearings that failed less than month later: forced tank opening and gas recovery
FROM CORONA NEEDLES TO RESISTORS AND TURBOPUMPED STRIPPER SYSTEM INSTALLATION
From corona needles to resistors

- 550 Mohm resistors ordered from NEC to replace some 60 corona gaps
  - Resistor based charge division change was relatively easy, except tight space
  - Lower voltages far more stable, accelerator has been run with 75kV at terminal

From corona needle based voltage division to resistors

2008
Turbopumped stripper change

- Original: N₂ gas stripper, extra gas pumped through HE- beam tube.
- Recirculation by turbo: About order of magnitude lower pressures at HE-side for same charge state distributions, even with larger holes at LE and HE terminal.
  - Less beam (charge state-) contamination due to residual stripper gas.

Turbo pumped stripper system 2012: higher transmission
BEAM LINES AND RECENT DEVELOPMENTS
The lab and the beam lines 2009
Low energy heavy ion ERDA

- Typically 1–20 MeV Cl, Br, I or Au ions from 1–3 MV tandem accelerator
- Time-of-flight–energy spectrometers for isotopic identification and energy spectrum measurement

Time of flight (velocity) and energy are measured for the same particle

\[ E = \frac{1}{2}mv^2 \Rightarrow m = 2E/v^2 \]

Different masses can be identified
Example: Thin film with high mass element

- Atomic layer deposited Ru film on HF cleaned Si
- Scattered beam, $^{35}\text{Cl}$, used for Ru depth profile
- Monte Carlo simulations needed for getting reliable values for light impurities at the middle of the film

Low energy heavy ion ERDA – See posters!

Poor E resolution
Gas ionization detector to replace Si-energy detector

- Why try to fix a well working system?
  - Greatly improved energy resolution for low energy heavy ions → heavier masses can be resolved
  - Gas detector is 1D position sensitive by nature → possibility for kinematic correction and therefore larger solid angles possible
  - Gas detector does not suffer from ion bombardment

Recoil ranges in isobutane

10.2 MeV $^{79}$Br 8.5 MeV $^{35}$Cl

Gas ionization detector development – See posters!
Gas detector performance

- Same **borosilicate sample** is measured with ToF-E, with two different energy detectors: a gas ionization detector with **thin SiN window** and new Si-detector.

**Beam: 10.2 MeV $^{79}$Br**

- **H = 290 keV**
- **B = 2.4 MeV**
- **Si = 4.5 MeV**
- **H, 10B, 11B, 16O, Na, Si, K, Ti, Fe, Zn, 79Br**

**Gas ionization detector with thin SiN window**

**New silicon detector**
‘Contaminants’ after analyzing magnet

- 13.6 MeV \(^{63}\text{Cu}^7\) CaPO (hydroxyapatite)

Is this mostly due to stripper gas effect in HE column?
Acknowledgements

TEKES-EU Regional Funds
Academy of Finland
TEKES

Accelerator based materials physics group at JYFL
Pelletron Laboratory

- 25 years old 1.7 MV Pelletron accelerator, in Jyväskylä since 2006
  - Available beams and energies: from H to Au, from 0.2 MeV to 20+ MeV

Three ion sources within ~2 m²!
Ion sources

- Mean life time of the ion source more important than 2 more beam
- Maintained by group, development together with ion source team

<table>
<thead>
<tr>
<th>Ion source name</th>
<th>Ion source type</th>
<th>primary ion(s)</th>
<th>Typical intensities</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alphatross</td>
<td>100 MHz RF, Rb charge exchange</td>
<td>He⁻</td>
<td>250 nA</td>
</tr>
<tr>
<td>SNICS '1'</td>
<td>Cs sputtering, single cathode</td>
<td>C⁻, Cl⁻, Cu⁻, Br⁻</td>
<td>2000 nA</td>
</tr>
<tr>
<td>PELLIS</td>
<td>Filament driven multicusp</td>
<td>H⁻</td>
<td>15 000 nA</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Ion source name</th>
<th>Typical operational parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>&quot;cathode&quot; HV, current</td>
</tr>
<tr>
<td></td>
<td>filament P, oven T</td>
</tr>
<tr>
<td></td>
<td>Einzel focusing HV</td>
</tr>
<tr>
<td></td>
<td>'Extraction' lens HV</td>
</tr>
<tr>
<td>Alphatross</td>
<td>+5 kV, 0.75 mA</td>
</tr>
<tr>
<td></td>
<td>250 °C / 55 °C</td>
</tr>
<tr>
<td></td>
<td>none</td>
</tr>
<tr>
<td></td>
<td>-8 kV</td>
</tr>
<tr>
<td>SNICS '1'</td>
<td>-4 kV, &lt; 0.2 mA</td>
</tr>
<tr>
<td></td>
<td>19 A, 6 V</td>
</tr>
<tr>
<td></td>
<td>-8 kV</td>
</tr>
<tr>
<td></td>
<td>-8 kV</td>
</tr>
<tr>
<td>PELLIS</td>
<td>+100 V, 1 A</td>
</tr>
<tr>
<td></td>
<td>70 A, 3 V</td>
</tr>
<tr>
<td></td>
<td>-5.5 and -3.4 kV</td>
</tr>
<tr>
<td></td>
<td>-10 kV</td>
</tr>
</tbody>
</table>
Where does the ions end up
Example: Thin film with high mass elements

- In ALD impurities from carrier gas or precursors may interfere in the process

**EXAMPLE:** LiTiO thin film, ~50 nm: Cl\(^{35}\) VS Br\(^{79}\)
- \(^{35}\)Cl close to perfect beam but cannot probe the Cl impurities in the film
- Heavier \(^{79}\)Br beam needed for this ’same as beam mass impurity’ search
- Br beam suffers from multiple scattering and cannot probe whole film

Low energy heavy ion ERDA
Lithography with light ions

- Ion beam lithography enables direct writing of deep '3D' structures, for example microfluidistic channels to the resists (PMMA) or even quartz.
- Stability in energy and fluence most critical, as for very small beam sizes the online monitoring is challenging.
- Uniform beam (up to ~30%), parallel exposure through slits → fast prototyping.
Lithography with light ions

- Ion beam lithography enables direct writing of deep '3D' structures, for example microfluidistic channels to the resists (PMMA) or even quartz.
- Stability in energy and fluence most critical, as for very small beam sizes the online monitoring is challenging.

Example which has OK parameters

Example with parameters NOT OK

Beam shift due to Alphatross sparking

Fluence change → not well developed
Need for negative helium

- Rutherford Backscattering Spectrometry (RBS) uses often He to probe the sample from the surface (few 10’s of nm to few µm).
- Higher the energy, lower the backscattering yield, but better mass resolution (and relative energy resolution at silicon detector).
- Lower the energy, better the depth resolution, but worse transmission through the accelerator → High He input current needed from ion source.

EXAMPLE: RBS for Pb that has diffused in glass at elevated temperatures

50 keV Pb implantation doses in glass
- 1E16 at. cm$^{-2}$
- 3E15 at. cm$^{-2}$
- 1E15 at. cm$^{-2}$
- 3E14 at. cm$^{-2}$
- 1E14 at. cm$^{-2}$
- 3E13 at. cm$^{-2}$

2 more doses!
What is needed: Conclusions I

- **ToF-ERDA**: Selection of heavy ions that are fast to switch, stability in intensity not that important but higher charge states from small accelerator is needed for reasonable count rates.

- **Lithography**: Stability in both energy and intensity most important. Light ions can create deep ’open’ structures where heavy ions can create closed channels directly (Bragg peak).

- **RBS**: Helium beam most used beam as it can separate heavy target/sample masses from each other

- **PIXE** (particle induced x-ray emission): Protons or helium most often used. Data can be collected often together with other methods easily.
What is needed: Conclusions II

- For ion beam applications, for both characterization and modification variety of negative ion beams is needed.

- Stable in energy and fluence, easy to maintain ion sources are priority parameters over higher beam intensities.

- **Protons:** PELLIS H\(^-\) source performs very well with long life times and is easy to operate by users.

- **Helium:** Alphatross currently has poor to worse performance. Upgrades coming: helical RF-plasma coupling and temperature stabilized Rb charge exchange chamber for stable Rb backflow to oven.

- **Heavy ions:** SNICS ’1’ has moderate performance. ”New” 40 MC-SNICS to be installed still in this year.
Growth of Al$_2$O$_3$ on TiO$_2$

- In dye sensitized solar cells even single ALD cycles of Al$_2$O$_3$ were found to reduce the interfacial electron transfer between semiconductor TiO$_2$ and dye molecule. This improves the performance of the cell.

- How thick films of Al$_2$O$_3$ were deposited during first cycles of ALD growth?

Liisa Antila, Mikko Heikkilä, Viivi Aumanen, Marianna Kemell, Pasi Myllyperkiö, Markku Leskelä, and Jouko E. I. Korppi-Tommola

Minimizing background

Sample: 50 nm TiO$_2$ on Si substrate where 5 ALD cycles of Al$_2$O$_3$ has been grow on to (corresponding 0.5 nm thickness)

All counts visible on log intensity scale

5 ALD cycles of Al$_2$O$_3$
Growth of Al$_2$O$_3$ on TiO$_2$

- Samples with 1, 2, 3 and 5 cycles of ALD Al$_2$O$_3$ on 50 nm ALD TiO$_2$ were studied
- Measurements using 8.26 MeV $^{79}$Br beam

Conclusion: Al$_2$O$_3$ film growth starts rapidly from the first cycles and then the growth rate per cycle reduces

![Graph showing Al area density vs ALD cycles](image)
Timing gates: construction

- **Carbon foils**
  - T1 foil 3 µg/cm² (diameter 9 mm)
  - T2 foil 10 µg/cm² (diameter 18 mm)
    - Determines solid angle of 0.29 msr
  - Distance between foils 633 mm

- **Wires in the electrostatic mirror grids**
  - Spot-welded 25 µm diameter Au wire
  - Wire-to-wire distance 1 mm, 97.5% transmission, telescope transmission 87%

- **Micro-channel plates**
  - Low-cost chevron-type MCP assembly from TECTRA
  - Active diameter >40 mm, pore size 12 µm, channel length/diameter 40:1
  - Specified only for 10 ns timing resolution!
TOF detection efficiency

- Detection efficiency against energy detector
- All sample elements, including H, can be detected and quantified
- No pinholes in C foils

![Graph showing detection efficiency against energy](typical ERDA energies)

**Detection efficiency (%)**
- Hydrogen
- Carbon
Timing resolution for He and H

- Current timing resolution 155 ps (FWHM) for 4.8 MeV incident He ions and 235 ps for 0.6 MeV incident H ions scattered from 1-2 nm Au film on SiO₂/Si substrate.

For He 155 ps timing resolution gives 32 keV resolution at 4.8 MeV but 0.8 keV resolution at 400 keV!
Example: Diamond-like carbon films

- 2.3 µm thick diamond-like-carbon film on Si, measured with 9 MeV $^{35}$Cl
- All isotopes can be determined for light masses
- Light elements can be well quantified (N content 0.05 ± 0.02 at.%)
Selection of coincident events

Time-stamp resolution 25 ns.

<table>
<thead>
<tr>
<th>ADC n:o</th>
<th>Ch</th>
<th>Timestamp</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>2675</td>
<td>457669383399</td>
</tr>
<tr>
<td>1</td>
<td>2624</td>
<td>457669383471</td>
</tr>
<tr>
<td>0</td>
<td>3756</td>
<td>457669459909</td>
</tr>
<tr>
<td>1</td>
<td>1332</td>
<td>457669459982</td>
</tr>
<tr>
<td>0</td>
<td>6688</td>
<td>457669499237</td>
</tr>
<tr>
<td>0</td>
<td>3044</td>
<td>457669862394</td>
</tr>
<tr>
<td>1</td>
<td>1987</td>
<td>457669862467</td>
</tr>
</tbody>
</table>

Lonely TOF-event, has probably hit T2 detector C-foil frame
Selection of coincident events

Coincidence <10µs, 585117 counts

1.6-1.9 µs, 576741 counts
1725-1750 ns, 329445 counts

outside peak, <10µs
ANALYSIS OF THE $\text{Al}_2\text{O}_3/\text{TiO}_2$ NANOLAMINATES
Measurements

- 9.9 MeV $^{35}\text{Cl}^{5+}$ - all samples, reflectance 69.5 geometry and 84
- Quick test to all samples with standard 1 MeV $^{4}\text{He}^{1+}$ RBS at 168

RBS - R2, 10 nm layers

ToF-E, R2, 10 nm layers
Measurements

- 6.0 MeV $^{12}$C$^{3+}$ - 3 thinnest layered samples, 84 and 86
- 0.5 MeV $^4$He$^{1+}$ - Rutherford Scattering to forward angles up to 88

Sample R3, 5 nm layers

Sample R4, 2 nm layers
Monte Carlo -simulations

- MC-simulations made for the spectra in reflectance geometry
- Better understanding of the composition and thicknesses

Sample R2, 10 nm layers, start profile

Difference comes from the incomplete selection of Al counts from Si substrate (does not effect MC-result)
FURTHER IMPROVEMENTS
Gas ionization detector

- Thin (~100 nm) SiN window
- Electrons for T2 timing signal emitted from the membrane
Conclusions

- All Al$_2$O$_3$/TiO$_2$ nanolaminates could be depth profiles and impurities, including hydrogen were analyzed.
- Nanolaminates with individual layers of 5 nm could be resolved.
- Depth resolution of <2 nm at the surface was reached.
- Gas ionization detector as an energy detector and yet coming position sensitivity will push the performance to even higher level.
Future improvements: Gas ionization detector

TOF-E results from ETH Zürich
Incident ion 12 MeV $^{127}$I and borosilicate glass target


200 nm thick SiN membrane from Aalto University, Finland, on 100 mm wafer
ALD 8.6 nm Al₂O₃/Si

- Atomic layer deposited Al₂O₃ film on silicon (Prof. Ritala, U. of Helsinki)
- Density of 2.9 g/cm³ and thickness of 8.6 nm determined with XRR (Ritala)
- Elemental concentrations in the film bulk as determined with TOF ERDA are O 60.3 at.%, Al 35.2 at.%, H 4.1 at.%, and C 0.5 0.2 at.%
10 nm CN$_x$ on silicon

- TOF-ERDA results from sputter deposited 10 nm thick CN$_x$ hard coating on Si. Measured with 6 MeV $^{35}$Cl beam and extreme glancing angle of 3°.
- A density of 2.0 g/cm$^3$ was used in converting areal densities to nm.
Effect of stripper gas pressure

- 13.6 MeV $^{63}\text{Cu}^{7+}$ CaPO (hydroxyapatite)
Timing resolution

- Shape of the MCP signal with the original anode
  - Risetime ~3 ns for He ions
Electronics and data acquisition

- **Timing gates**
  - Fast Phillips Scientific 776 preamplifier (10)
  - Ortec 935 quad CFD
  - FAST 7072T TDC/ADC, no delay used

- **Energy detector**
  - Implanted 450 mm² ORTEC detector (ULTRA series)
  - Ortec 142 preamplifier
  - Ortec 571 amplifier
  - FAST 7072T TDC/ADC

- **Data acquisition with LabVIEW**
  - National Instruments FPGA card based multiparameter data acquisition system, programmed in Jyväskylä
  - 40 MHz (25 ns) time stamping
  - Can host up to 8 ADCs, easily expandable
  - List-mode data – coincident events determined off-line!
Depth resolution optimization

\( ^{35}\text{Cl} \Rightarrow \text{Si} \)

oxygen recoils
angles 5.0°, 38.3°

Depth resolution (nm)

Beam energy (MeV)
TOF-ERDA beamline and chamber

- UHV compatible chamber with a load lock
- LabVIEW controlled stepping motor driven 6-axis goniometer (Panmure instruments)
- Currently sample holder for two samples, holder for 7 samples in design
- Beamline equipped with high precision slits and NEC beam profile monitor
- Telescope angle 41°
Timing gates: voltages

MCP stack and "toblerone"

- HV2: -1800 V
- 10 kΩ
- 1699 V
- 20 MΩ
- 99 V
- 1 MΩ

Carbon foil and outer mirror grid

Cable from anode to SMA leadthrough
Towards position sensitivity

- Risetime of 1 ns with PCB anode achieved with the test detector
- Risetime of 2 ns from lower MCP electrode through 1 nF capacitor achieved with the test detector

Also ALD-Al$_2$O$_3$ coated carbon foils have been fabricated, much higher electron yields expected
Conclusions

- New high performance spectrometer has been built in Jyväskylä
- Detector telescope has high detection efficiency and good timing resolution
- Depth resolution of <2 nm at the surface has been reached
- Position sensitivity and gas ionization detector as an energy detector will push the performance to even higher level

1st timing detector, 3 μg/cm² C-foil
2nd timing detector, 10 μg/cm² C-foil