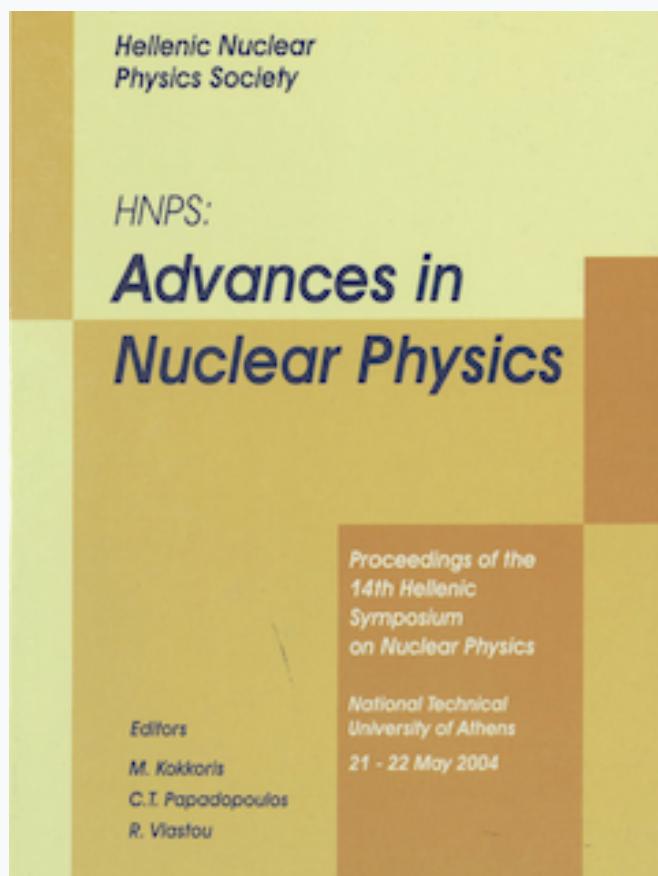


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# Applied Nuclear Physics-Advanced Ion Beam Materials Analysis at small Accelerators

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## Abstract

The Rossendorf Ion Beam Laboratory has been developed to an international large scale user facility in the field of ion beam physics and ion beam materials research. The laboratory operates a large number of modern experimental equipment at three MeV accelerators, three implanters, an ECR source and a FIB which together provide almost all ions species in a wide energy range from a few hundred eV to a few ten MeV. Also IBAD and PIII devices were installed for various purposes. The research stations at the accelerators are supplemented by complementary techniques like TEM, SEM, AUGER, AFM etc., all contributing useful information to thin film investigations. In this paper a short overview of the laboratory is given and a few recent experiments and their results are shown. Rutherford Backscattering Spectrometry (RBS) and Elastic Recoil Detection Analysis (ERDA) are well established techniques for quantitative thin film. The advantage of these methods consists in the simple physics they are basing on, namely the stopping of energetic ions in matter and the binary scattering of at the Coulomb potential of atomic nuclei. The increasing importance of ultra-thin layers for novel technologies demands quantitative analysis techniques with a depth resolution of atomic monolayers, which can be obtained for RBS and ERDA by magnetic spectrometers only. The magnetic spectrometers we have installed at the 3 MV Tandetron and at the 5 MV tandem are described, recent applications are shown and a few problems to achieve high depth resolution will be discussed. Heavy ion detectors as Bragg IC, dE-Erest-telescopes and ToF spectrometers, developed for nuclear physics experiments, are now applied for ERDA, providing an efficient analysis of thin films containing light elements. The lateral position resolution of such detectors enables kinematic corrections and allows large solid angles. Thus by ERDA in situ studies during surface modification processes are possible like in the case of the nitridation of aluminum and stainless steel. At the external beam mainly objects of fine arts or of historical value are analysed. It will be shown, how the complementary application of PIXE, RBS and PIGE can help to detect the beginning corrosion of mediaeval glass objects.

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## 1 Introduction

Ion beam materials analysis (IBA) at MeV accelerators is not a new technique. Rutherford backscattering spectrometry (RBS) , introduced about 40 years ago, became a standard method in today's toolbox for thin film analysis. Both in its concept and in its execution, RBS is quite a simple experiment and used widely in the past to measure the elemental composition of thin films.

Indeed experimental nuclear physicists long before used scattering and nuclear reactions to determine the composition of their targets and monitor it during the experiment.

One of the reasons of the broad application of RBS at the end of the 60's was that many of small van de Graaff accelerators were not longer interesting for nuclear physicists and became available for other kind of experiments. Another reason was the availability of simple and cheap solid state detectors with energy resolutions well below 1 % , together with the technical development of easy-to-use multichannel analysers.

The strength of RBS consists in the very simple physics, the scattering of the light incidence ion nuclei at the Coulomb potential of the target nuclei in elastic binary collisions. The kinematics and the Z2-dependence of the Rutherford cross section cause however a drawback of this method ; it is almost blind for light elements in a heavier matrix.

Therefore not much later then the introduction of RBS the spectrometry of the recoil atoms, ejected from the target after collisions with heavy incidence ions , was used for depth profiling of light elements in materials. At its early days in this analysis technique (ERDA) solid state detectors were used, with a thin foil in front to stop elastically scattered heavy ions. Later on more sophisticated detectors , developed in the field of nuclear physics, were applied, which provide not only an energy signal but enable particle identification and position readout.

Besides of the advantage of an easy quantification of IBA results there is another feature which attracts an increasing interest. Beams of light MeV ions can penetrate through thin foils, separating the vacuum of the beam line of the accelerator from the atmosphere. In air the range of these ions is in the order of some 100 mm. Thus near the exit foil all kind of subjects can be placed for ion beam irradiation., which cannot be brought into vacuum.. This allows for instance materials analysis of historical objects of fine arts like paintings, glass, porcelain etc. Here besides of RBS mainly the excitation of characteristic X-rays (PIXE) and  $\gamma$ -quanta (PIGE) is employed.

In particular the shrinking dimensions in many branches of contemporary

technology demand characterization of ultrathin films with a depth resolution below 1 nm, which can be obtained by high energy resolution  $\Delta E/E$  of 10-3 in RBS or ERDA. Here again IBA benefits from the early spectrometer developments in nuclear physics. About 50 years ago Browne and Buechner at MIT described a simple magnetic spectrometer with circular pole shapes. Many copies of this instruments were used as workhorses at accelerators around the world. Later on more sophisticated spectrometers were constructed and used, but mostly decommissioned before the ion beam analysts became aware of their unique virtues. Only the Q3D spectrometer at the Munich tandem accelerator is used with great success for high resolution ERDA [1].

In the following a few examples are given, how these technical developments have been implemented at the ion beam laboratory of the Forschungszentrum Rossendorf in order to provide IBA of advanced materials.

## 2 RBS with high depth resolution

At our ion beam laboratory two magnetic spectrometers and one electrostatic analyser (ESA) are used for high resolution ion beam analysis and ion-solid interaction studies.

A small magnetic spectrometer of Browne-Buechner type was installed for HRBS and NRA at the beamline crossing of the 500 kV implanter and the 3 MV Tandetron. At this position beams in a wide range of ion species with energies from about 50 keV to 15 MeV are available.

The spectrometer consists of an UHV scattering chamber equipped with facilities for sample treatment and characterisation like sputter cleaning, low-rate UHV e-beam evaporators, residual gas analyser and a RHEED system, of the single dipole magnet with circular pole pieces and of the focal plane detector. Details of the set-up and the parameters of the spectrometer are given in [2].

At the 5 MV tandem accelerator the QQDS magnetic spectrometer "Little John" is being commissioned for HRBS, HERDA and experiments in transmission geometry. To achieve a variable dispersion the focal plane detector can be placed at different positions [3].

The spectrometer can be rotated around the axis of the 600 mm UHV scattering chamber. This allows detection angle settings of  $0^\circ$ ,  $15^\circ$ ,  $30^\circ$ ,  $45^\circ$  and  $60^\circ$ . The parameters of this spectrometer are given in Table 1.

The electrostatic ion analyser with a maximum energy-charge product of 4 MeV\* q is mainly used for measurements of energy loss and charge state

distributions in transmission geometry [4] or as a high resolution energy filter for heavy ions to study the response functions of various heavy ion detectors [5].

The scattering chambers of both spectrometers are similar and allow various in- situ sample treatments. The chambers are equipped with precision UHV five-axis-goniometers, the prerequisite for channelling/blocking experiments. Samples are transferred with a load lock system to docking stations at the goniometer, where the heater and the thermocouple are mounted.

For experiments requiring very high sample temperatures a special 2-axis goniometer with virtual tilt axis, consisting of a differentially pumped UHV rotational platform, a  $\pm 5^\circ$  tilt stage and linear stage, has been constructed. A electron beam heater provide sample temperatures up to 2000  $^\circ$ C.

Fig. 1 demonstrates the achievement potential of the Browne-Buechner spectrometer. The upper part shows a TEM image of a multilayer  $30 \times (1.9 \text{ nm Mo} / 2.33 \text{ nm B}_4\text{C})$  on Si demonstrating the high regularity of the alternating layers. The lower part shows a spectrum obtained with 2 MeV C ions at a scattering angle of  $35.5^\circ$  with an incidence angle of  $17.5^\circ$  with respect to the surface. The line is a spectrum fit considering straggling and interface roughness [6].

### 3 A novel ERDA detector , a combination of dE-E gas ionization chamber and Time-of-Flight spectrometer

A large acceptance angle, kinematical correction, sufficient energy resolution and good particle separation are desired features for Heavy Ion ERDA. Most of the TOF – Energy – Telescopes have a good mass separation but a small acceptance angle due to the long flight path and the small size of energy detectors. Furthermore the mostly used Si-detectors suffer from the radiation damage and have an unacceptable energy resolution for heavy ion detection. Therefore we combined a dE-E gas ionisation chamber (IC) with an  $52 \text{ cm}^2$  entrance window, which corresponds to an acceptance angle of  $3.1 \text{ msr}$  as energy detector with a time-of-flight spectrometer in front of. The chamber is position sensitive in the scattering plane and enables a kinematical correction of the energy signals with an angle resolution of  $\pm 0.1^\circ$ . A small MCP detector with a thin Carbon foil tilted by  $45^\circ$  gives the start signal near the target. A large MCP detector with a nearly perpendicular Carbon foil and electrostatic mirror just in front of the entrance window of the IC delivers the stop signal. Optionally the particle separation can be derived from the ToF- or from the dE/E signal ratio.

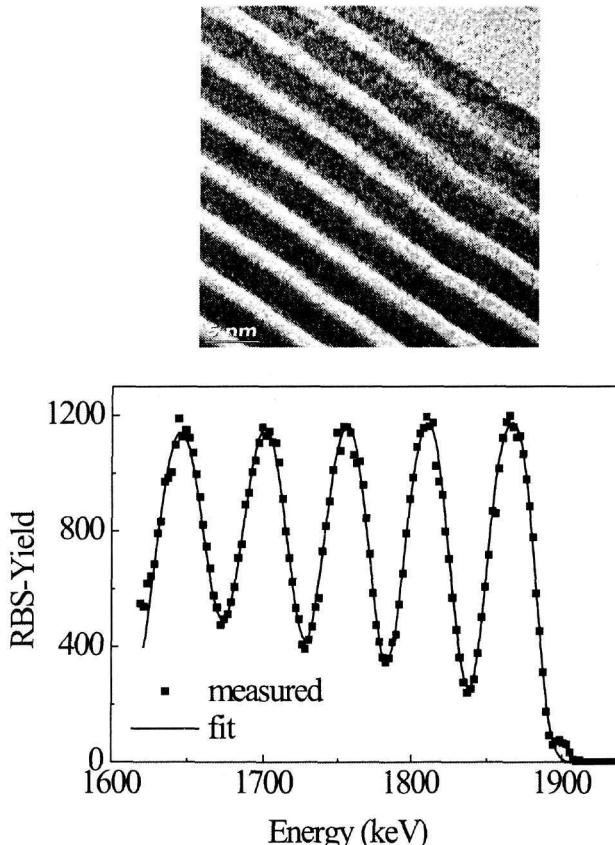


Fig. 1. Above: TEM-image of a Mo/B<sub>4</sub>C multilayer on Si. The dark layers show Mo, while the bright layers show B<sub>4</sub>C. Below: Spectrum of this sample as measured with 2 MeV C-ions at an incidence angle of 17.5° and a scattering angle of 35.5°.

Fig. 2 shows the schematic setup and the installation at the ERDA chamber.

In Fig. 3 the 2-D plot of an ERDA spectrum of an TiO<sub>2</sub> / SiO<sub>2</sub> layer stack of an optical Filter is displayed.

#### 4 External proton beam

The proton beam on atmosphere allows non-destructive studies on unique objects. In particular, pieces of art can be analysed without sampling or any visible damage.

In the Rossendorf layout the proton beam of 4 MeV energy penetrates a

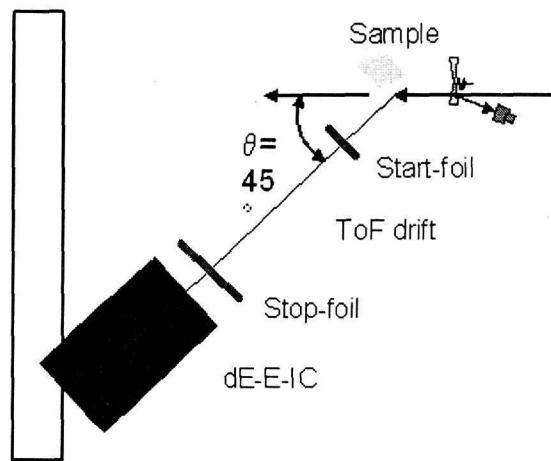


Fig. 2.

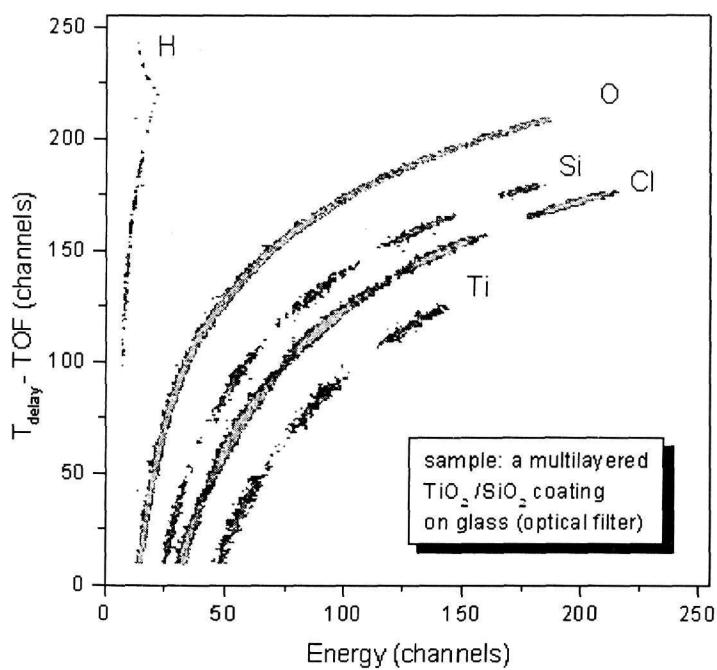


Fig. 3.

HAVAR<sup>®</sup> foil of 2  $\mu\text{m}$  thickness (energy loss about 100 keV) and it is guided on atmospheric pressure within a 3 l/min gas flow of helium. The latter reduces proton stopping as well as X-ray absorption and it prevents the emission of Ar K-radiation from air. The common ion beam techniques PIXE, RBS and PIGE are implemented for simultaneous measurements. Thus, sensitive materials like paper or white porcelain can be examined by a single proton irradiation. The Rossendorf external beam set-up joins two Si(Li) detectors, namely PIXE1: active area of 12  $\text{mm}^2$  - no filter, and PIXE2: 80  $\text{mm}^2$  - 1 mm acrylic absorber. This PIXE arrangement allows detection of elements with both lower and higher atomic number in an effective manner. Analysis of selected light elements, i.e. B, Na, Mg, Al, Si, takes place by PIGE using a large volume HPGe detector of 60% efficiency. Backscattered protons are applied for surface characterisation of the object using a light protected and cleanable surface barrier detector of 100  $\text{mm}^2$  area. The detectors are arranged at backward angles. They are positioned close to the beam spot (distances  $\sim$  30 mm, HpGe  $\sim$  80 mm) in order to obtain large solid angles. Thus, sensitive materials can be studied using low proton beam currents ( $\sim$  200 pA) and short acquisition times ( $\leq$  1 min).

Typical objects have been easel paintings and hand drawings, book paintings, porcelain painting, but also full quantitative analysis of glass, enamel and gold. The usual subject of analytical work was the understanding of individual artistic technologies. In particular, depth resolved information has been possible using RBS (surface characterization) and PIXE at varied proton energy (e.g. detection of paint layers). As an example Fig. 4 shows PIXE and RBS spectra obtained from two golden details of a reverse painting on glass [7].

As revealed by PIXE both the golden frame and the golden monogram proved to be made from brass of one and the same material, see PIXE: the intensity ratio Cu/Zn. Whereas the frame consists of a brass foil, the broad C signal arises from the sticking material, the monogram was painted - Pb comes from lead white certainly admixed with the brass powder.

## 5 Conclusions

All the techniques of ion beam materials analysis has been established as valuable and often indispensable analytical tools for thin film characterisation. At the ion beam laboratory of the Forschungszentrum Rossendorf advanced experimental facilities for these techniques are installed at three accelerators. As a user facility at an European scale all the facilities are available to guests and scientists are given the possibility to conduct their own research at this laboratory.

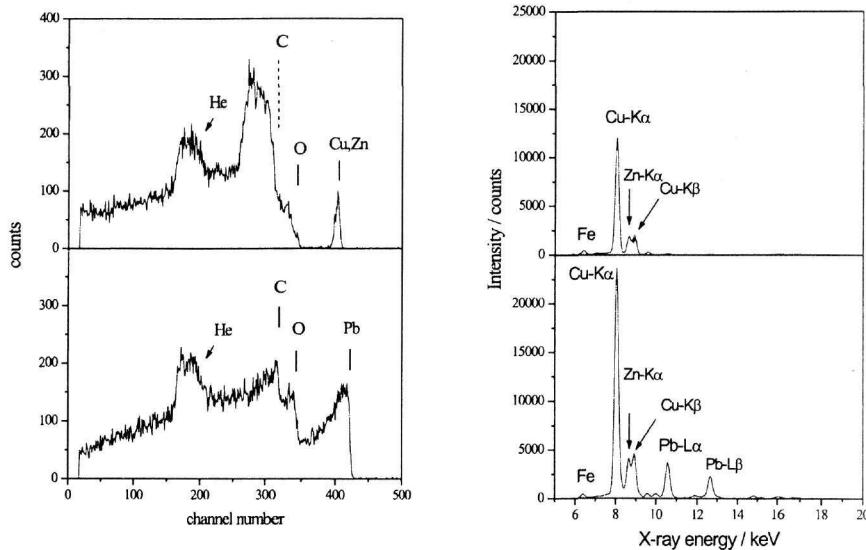


Fig. 4. RBS: above - brass foil below - painted monogram PIXE: above - brass foil below - painted monogram

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