Elastic recoil detection using heavy ion beams

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SUMMARY Elastic Recoil Detection using heavy ion projectile beams allows compositional depth-profiling of materials to a depth of about 2.5 μm. The technique is sensitive to all chemical elements including hydrogen. It is particularly suited for the analysis of thin film materials. Large solid-angle position-sensitive gas ionization detectors have been developed for the efficient detection of the recoil ions. With the set-up at the Australian National University, measurement and analysis are greatly simplified by using a new detector design. A grid electrode allows a direct determination of the ion energy, while a divided anode enables the simultaneous detection of ions with largely different atomic numbers and also provides linear position information. A diverse spectrum of materials has been analyzed including photosensitive doped silica, high-T_c superconductors and dielectric films.

1. Introduction

Heavy ion beams produced by electrostatic tandem accelerators such as the 14 UD Pelletron at the Australian National University can be utilized to depth-profile the composition of material surfaces and thin films using the Elastic Recoil Detection (ERD) technique (1). During a brief exposure of the sample to the beam, employing a glancing geometry, the incident beam particles eject, in elastic collisions, recoil ions out of the sample. These ions are detected and distinguished according to their atomic number. Since the energy of the ions is related to the depth inside the sample from where they originate, the yields detected for different ion species accurately reflect the material composition as a function of depth. Thus the total number of atoms of a chemical element present at a certain depth can directly be inferred from the measurement. The energy-depth relation is established using the stopping power of the material for the incident and the recoiling ion.

In order to increase detection efficiency and minimize beam related sample modifications, large solid-angle (~ 5msr) position-sensitive gas ionization detectors have been developed (2). With such detectors the atomic number of the ions is determined from their initial energy loss inside the detector. Total energy information is obtained by collecting all ionization electrons. The position information allows the correction of kinematic energy differences of ions from the same depth, but with different scattering angles. Thus the large acceptance angle can be reconciled with optimum energy resolution. In contrast to solid state detectors, gas ionization detectors are not degraded by ion damage.

All chemical elements, including hydrogen, can be detected simultaneously, with similar sensitivity and depth-resolution. Elemental separation is best for the lighter elements, with neighbouring elements being fully

resolved for atomic numbers up to about $Z=20$.
Concentrations of ~ 0.1 atomic percent can be detected. The sampling depth depends on the sample material and is of the order of 1.5-2.5 μm. For the surface region a depth-resolution of 10 nm can be achieved. It deteriorates, however, with increasing depth because of the energy straggling and multi-scattering of the ions in the sample.

The ERD detector (3-5) at the Australian National University includes several new design features which greatly simplify detection and analysis. Among those are *the subdivision of the energy loss electrode*, which allows the simultaneous identification of heavy and lighter ions at the same detector gas pressure, *a novel grid electrode* for the total energy measurement, which obviates the need for relative calibrations of electrodes, and *a saw-tooth electrode within the anode*, which provides linear position information.
The applications of the technique are diverse, with thin film analysis being a particular strength. Among the thin film materials analyzed at the Australian National University were photo-sensitive doped silica films for opto-electronic applications, magnetically sensitive films for data storage, dielectric films, such as silicon nitride and tantalum oxide, hydrogenated carbon films and high-T_c superconducting films. Other applications included nitrided stainless steels, where nitrogen and hydrogen depth-profiles were of particular interest, and the measurement of oxygen depth-profiles in high-T_c superconductors.

2. Experimental Set-up

Figure 1 shows the detection geometry.

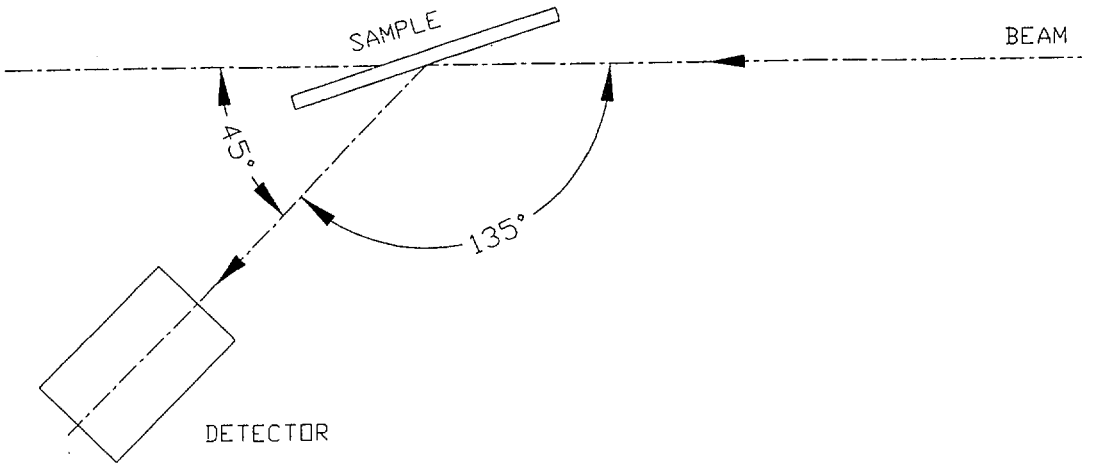

Figure 1. Schematic diagram of the detection geometry.

Typical projectile beams are gold and silver, at beam energies greater than 1 MeV/amu. The detector is mounted on a movable arm inside a large scattering chamber with a diameter of 2 m. The detection angle can be

varied continuously between 30° – 60°. The distance between detector and sample is 28 cm. The vacuum inside the scattering chamber is ~10^{-7} mbar. The sample holder accommodates six samples and can be rotated to vary the angle between the incident beam and the sample surface. The samples are loaded through a vacuum lock.

Propane gas is passed through the detector at a constant pressure in the range 40 - 100 mbar, which is chosen to stop the lightest ions. Ions recoiling from the sample enter the detector through a grid-supported mylar window which is 0.5 μm thick. The detector is collimated with a circular aperture (diameter 20 mm).

A schematic diagram of the detector interior is shown in Figure 2. The electrode-stack consists of a cathode, a Frisch grid, a grid electrode and an anode. The anode is divided into two ΔE sections (ΔE_1 and ΔE_2) and a residual energy section (E_{res}). The middle section (ΔE_2) is subdivided using a saw-tooth geometry. Signals are obtained from all anode sections, the grid electrode and the cathode.

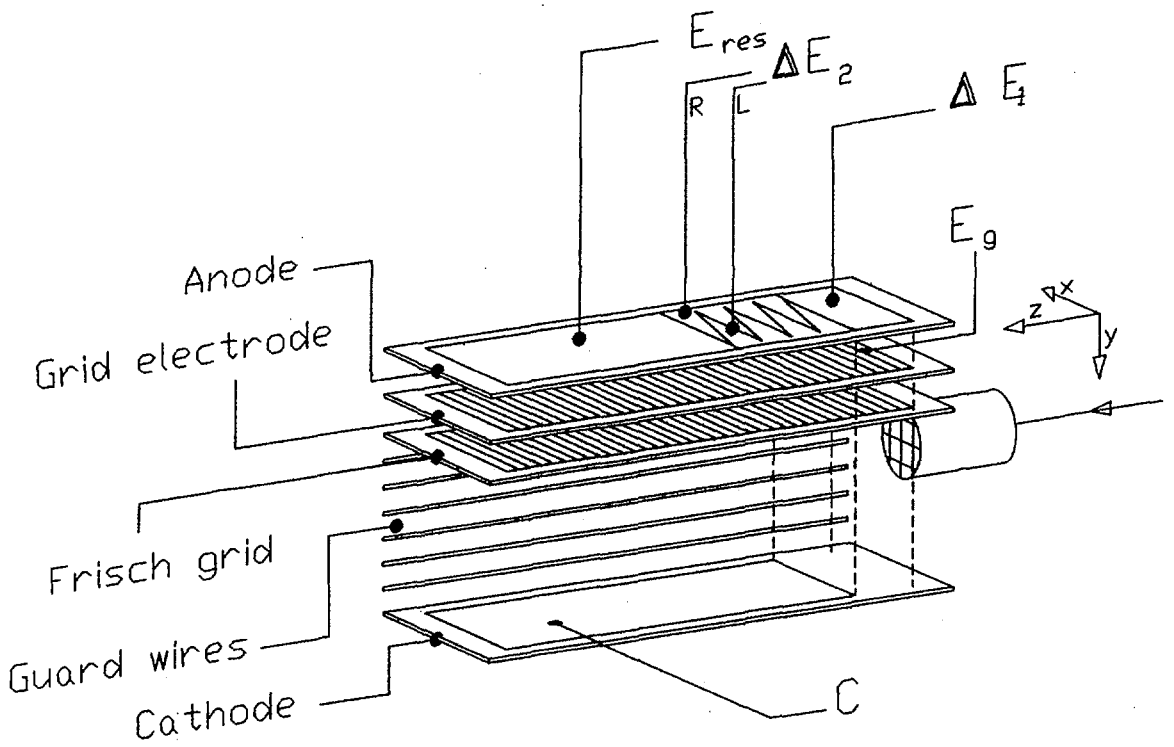

Figure 2. The new detector design used at the Australian National University.

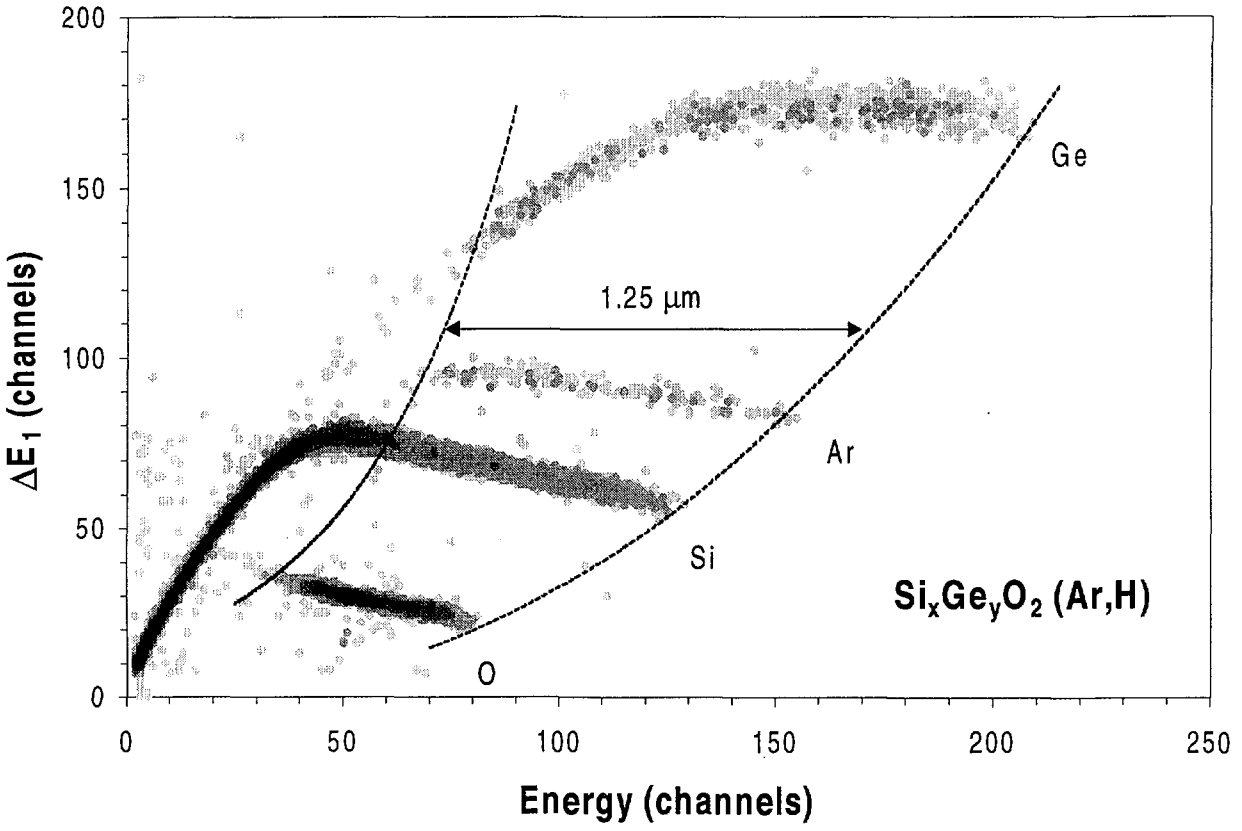

Figure 3. A two-dimensional spectrum of the ΔE_1 and energy signals detected for recoils from a Ge-doped silica film. The hydrogen content is detected simultaneously (not shown here).

Total energy information is either obtained by combining the signals from the anode, or directly from the grid electrode. The signals ΔE_1 and ΔE_2 are proportional to the initial energy loss rate of the recoil ions in the detector, which is a function of the atomic number Z, and allow the separation of different ion species.

The horizontal position of the ion track is determined by combining the amplitudes of the signals from the right (R) and left (L) saw-tooth electrodes according to (R-L)/(R+L). The vertical position of the ion track is obtained from the ratio of the cathode signal and the sum of ΔE_2 and E_{res}. With this position information and given detector angle and detector distance, the scattering angle θ can be determined.

The energy E of the recoiling ions changes with θ according to

$$E = \left[4m_p m_r / (m_p + m_r)^2\right] E_p \cos^2\theta$$

where E_p is the projectile energy and m_p and m_r are the masses of projectile and recoil ion, respectively. The kinematic energy difference of the ions over the detector acceptance is corrected with

$$E^{cor} = E \frac{\cos^2\theta_0}{\cos^2\theta}$$

where θ_0 is the detector angle and E^{cor} the corrected energy.

3. Photosensitive doped-silica films

The capabilities of the technique may be demonstrated using the example of photosensitive germanium- and tin-doped silica films. These materials are being developed for integrated photonics applications. Such films can be deposited by plasma assisted deposition techniques and their initial refractive index and photosensitivity are determined by the dopant:Si:O stoichiometry. The presence of hydrogen in the film is deterimental to their performance, because it causes optical absorption in the 1.3-1.5 μm wavelength range of interest for telecommunications. Characterization of the films therefore requires accurate determination of the film composition, including the presence of hydrogen (5). Figure 3 shows a plot of the ΔE_1 signal as a function of the total energy signal, as detected for recoil ions from a germanium-doped film. The contours represent the detected ion yield. Germanium, argon (present as a contaminant), silicon and oxygen can be identified. The film limits are indicated. The stoichiometry can be extracted from the total number of recoils from within the film, after normalisation with the scattering cross sections. This assumes that all elements are distibuted uniformly throughout the film. The film uniformity can be assessed when individual energy spectra are extracted from the two-dimensional projection of ΔE_1 versus energy. This is shown in Figure 4.

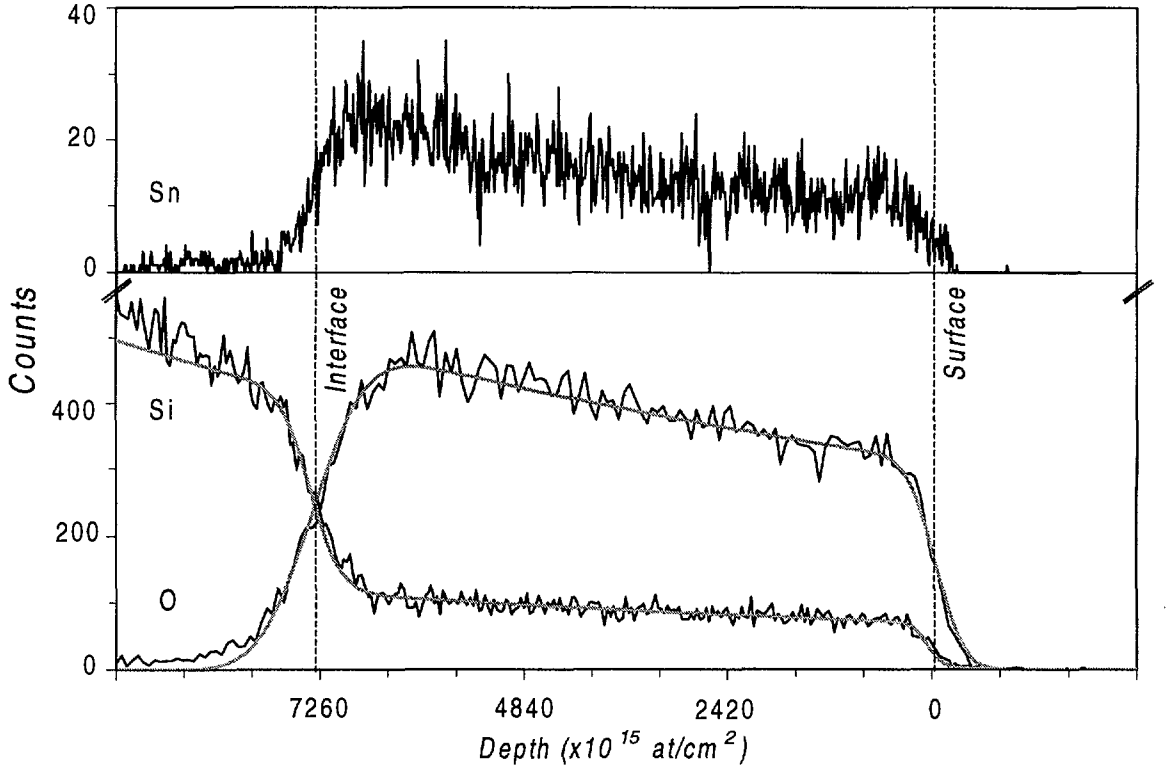

Figure 4. Energy spectra obtained for a tin-doped silica film. The spectra in the lower part of the figure are compared with simulations assuming uniform stoichiometry.

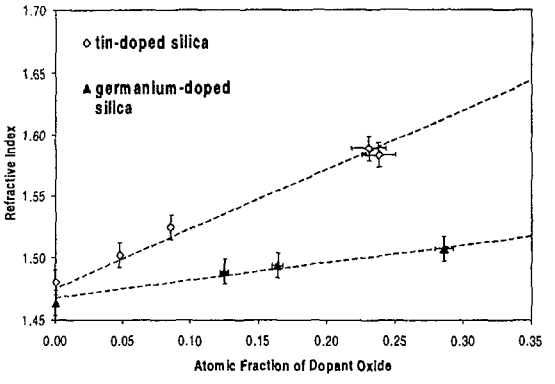

Figure 5. The refractive index of doped silica films plotted as a function of the atomic fraction of the dopant oxide as measured with heavy ion ERD.

The refractive indices of the films analyzed are plotted in Figure 5 as a function of the measured dopant fraction. Both sets of data exhibit a linear behaviour with different slopes.

4. Summary

Elastic Recoil Detection (ERD) using heavy ion beams can be used to depth-profile the composition of materials. It is particularly suited for thin film analysis. At the Australian National University a new large solid-angle gas ionization detector with position sensitivity is used to detect the recoil ions efficiently and with a minimum of calibrations. The capabilities of the system have been demonstrated using the example of photosensitive doped silica films.

5. References

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