Nuclear Instruments and Methods in Physics Research B 273 (2012) 6–10

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

Heavy ion energy loss straggling data from Time of Flight stopping force measurements

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ARTICLE INFO

Article history:
Available online 4 August 2011

Keywords:
Stopping force
Energy loss straggling
Heavy ions
ToF-ERDA

ABSTRACT

The accuracy of heavy ion beam analytical techniques such as Heavy Ion-Elastic Recoil Detection Analysis (HI-ERDA) depends on, among other factors, the accuracy of basic ion beam data such as stopping force and energy loss straggling, used as input in ion beam analysis programs. The adaptation of the Time of Flight (ToF) ERDA technique for stopping force measurements in free standing target foils brings with it the possibility of extracting energy loss straggling information from the ToF derived energy spectra. In essence several straggling values can be obtained over a continuous energy range from a single measurement. A presentation is made here of an exploration of that possibility using the passage of ^{12}C, ^{16}O, ^{27}Al and ^{84}Kr ions in the oxide ceramic ZrO_2 as examples. The experimental straggling values, obtained over the 0.1–0.6 MeV/u energy range, are normalised to Bohr straggling and compared with Yang's empirical formulation, which is widely used in ion beam analysis programs.

1. Introduction

The study of the passage of energetic ions through matter is now a well established field that continues to attract the attention of many researchers [1–8]. This prolonged interest lends itself partly to the need to better understand and describe ion beam–matter interactions at the fundamental level, and equally important, to improve the quantitative accuracy of nuclear techniques in a range of ion beam applications. In ion beam analysis, for example, accurate stopping force and energy loss straggling values are of vital importance in the analysis and depth profiling of near-surface regions of thin film structures.

When a mono-energetic (∼MeV) ion beam passes through matter it slows down mainly due to Coulomb interactions with target electrons. This energy loss process is subject to statistical fluctuations and as a result there is a spread in the emerging beam energy distribution. The energy spread is termed electronic energy loss straggling and for many practical purposes, the energy distribution can be modelled by a Gaussian and so can be characterised by the average square fluctuation in energy loss [2]. There are several theoretical formulations that describe electronic energy loss in matter [3], and while there is still scope for further refinement, some of the semi-empirical models [4,5] have over the years been largely accepted as sufficient for most purposes. There is also a relatively substantial amount of experimental stopping force data for light ions (H, He) found in the literature spanning several decades to validate these predictive formulations. The amount of data for heavy ions, while still far from adequate, is also gradually increasing. The latter development has been buoyed not in the least by the need for accurate ion beam data for heavy ion depth profiling of thin films.

The above state of affairs is quite different when it comes to straggling data; theoretical predictions of energy loss straggling are not well proven, particularly for heavy ions near the stopping force maximum [6] and experimental straggling data is even rarer for heavy ions. Besenbacher and coworkers [2] chronicle some of the earliest attempts at predicting electronic energy loss straggling. These began with Bohr's theory based on the interaction between a fully stripped fast ion and a free electron gas. Bohr in 1915 used a classical model to derive a simple expression to estimate straggling. For a layer of thickness Δx, Bohr straggling Ω_B is given by

$$\Omega_B^2 = 4\pi(Z_1 e^2)^2 N Z_2 \Delta x \quad (1)$$

where Z_1, Z_2 are the projectile and target ion atomic numbers, respectively and N is the atomic density of the target material. Lindhard and Scharff later extended Bohr's treatment by considering an atomic electron density that distinguishes between the inner and outer electrons of the electron cloud. Further on Bonderup and Hvelplund improved the Lindhard–Scharff model by applying the Lenz–Jensen model for the atomic electron density. Using the

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doi:10.1016/j.nimb.2011.07.024

Lenz–Jensen model is believed to lead to a more correct description of straggling as it takes into account contribution from different parts of the electron cloud, in particular target electrons bunched into atoms or molecules [6]. Chu used an alternative electron density model, the Hartree–Fork–Slatter model, to modify Bohr's classical formula by taking into account deviations caused by electron binding in target atoms [7]. Yang et al. [7] derived an empirical formula based on Chu's theory. Their procedure (commonly referred to as Yang straggling) takes into account charge state fluctuations and correlation effects, and is summed up by the following expression for energy loss straggling;

$$\frac{\Omega^2_{Yang}}{\Omega^2_B} = \gamma^2(Z_1, Z_2, v)\frac{\Omega^2_{Chu}}{\Omega^2_B} + \frac{\Delta\Omega^2}{\Omega^2_B} \qquad (2)$$

where $\gamma(Z_1, Z_2, v)$ is the effective charge factor for ions in matter, v the projectile ion velocity, and $\Delta\Omega^2$ is the additional straggling due to correlation effects.

Yang straggling is the most widely used electronic energy loss straggling model in modern ion beam analysis codes such as SIMNRA, NDF, CORTEO and MCERD [9]. On the whole, the accuracy of the model particularly for heavy ions remains untested. It is against this background that this article seeks to introduce a method that can generate a range of experimental straggling values from a single measurement to add to the scant global database of heavy ion straggling data.

2. Experimental set up

Straggling data of heavy ions in ZrO_2 presented here is extracted from energy loss measurements of heavy ions through zirconia foils, performed using a Time of Flight-Elastic Recoil Detection (ToF-ERD) spectrometer. The basic experimental set up, detailed in an earlier publication [10] is illustrated in the sketch in Fig. 1. It is worth pointing out here that the orientation of the timing detectors T1 and T2 shown here is a more correct representation of the actual set up than reported earlier. A 27.5 MeV ^{84}Kr projectile ion beam was used to recoil ^{12}C, ^{16}O and ^{27}Al ions from suitable target samples (C-graphite, Al_2O_3) into the ToF telescope. For ^{84}Kr ions, a thick Au/Si target layer was used to forward scatter the incident ^{84}Kr beam into the detector system.

The energy E_1 of the recoil/scattered species before passing through the target stopper foil is determined from the measured ToF between the timing detectors T1 and T2, and the exit energy E_2, tagged by the Si surface barrier detector (SBD) is determined from a coincident ToF obtained in a second run *without* the stopper foil. The second run is, in essence a one-to-one energy calibration of the SBD for each energy bin for each ion species. The coincidence measurement of the ToF and the exit energy results in raw data in the form of 2-D ToF-E scatter plots. Fig 2 shows one such plot from the measurement of the stopping of ^{12}C recoils through one of the ZrO_2 foils.

The zirconia foils, produced by ACF Metals™, were characterised for roughness by Atomic Force Microscopy and Rutherford Backscattering Spectrometry (RBS) was used to measure the foil thickness and to confirm the stoichiometry. Two foils were used in two separate experiments. For ^{12}C, ^{16}O and ^{27}Al ions the thickness of the foil used was 331 ± 14 μg cm^{-2}, and for ^{84}Kr ions a foil of 345 ± 15 μg cm^{-2} was used.

2.1. Data analysis

The C → ZrO_2 example in Fig. 2 will be used to illustrate the data analysis procedure adopted here. Time of Flight (t_i) tagged by a particular SBD energy response is extracted from the 2-D ToF-E scatter plot by projecting onto the ToF axis the section (or slice) of the scatter plot corresponding to that energy. [The energy bin (or slice) width in the case of carbon recoils is about 30 keV, which is well below the ~5% energy resolution of the Si detector used here. The energy resolution for O and Al ions was also of the same magnitude, and for Kr ions it was slightly higher at ~9%.] In Fig. 2, t_1 and t_2 are the measured flight times with and without the stopper foil respectively and the mean peak values are obtained by fitting the projected time spectra with Gauss functions. Through time calibration t_1 is calculated to be 58.41 ns and t_2 is 65.56 ns, for a flight length L of 0.584 m [10].

The energy loss ΔE of a recoil (or scattered) ion of mass m after passing through the stopper foil is given by

$$\Delta E = E_1 - E_2 = \frac{m}{2}\left(\left(\frac{L}{t_1}\right)^2 - \left(\frac{L}{t_2}\right)^2\right) \qquad (3)$$

where E_1 and E_2 are the incident and exit energies, respectively. For the given times, the energy loss through a 0.331 mg cm^{-2} ZrO_2 foil works out to be 1.28 MeV at an average energy of 5.57 MeV. This gives a stopping power value of 3.9 MeV/mg cm^{-2}, which compares well with the SRIM predicted value of 4.1 MeV/mg cm^{-2} [4]. Full results of the stopping force evaluations can be found in Ref. [11].

For straggling evaluation the full width at half maximum (Ω-fwhm) of the energy distribution is in each case calculated from

$$\Omega_i = \frac{2\Delta t_i}{t_i}E_i \quad i = 1, 2 \qquad (4)$$

where Δt_i is the (fwhm) of the ToF projection. Whereas in traditional energy loss measurement experiments the incident energy is well defined and the exit energy is the unknown measurand, in

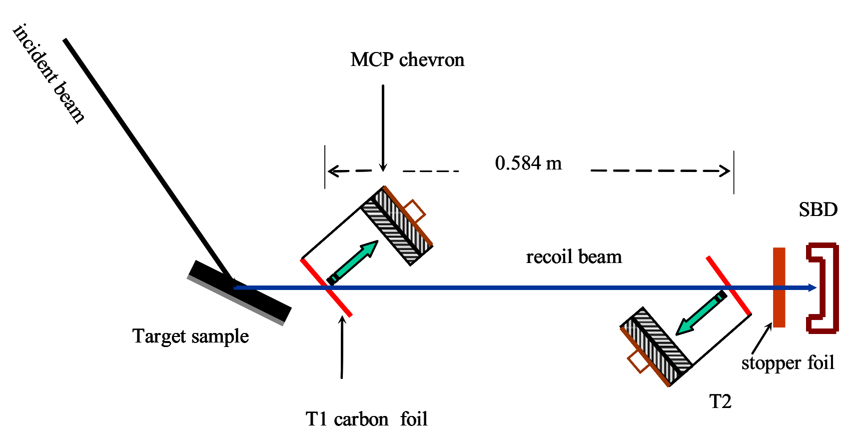

Fig. 1. A schematic illustrating set up of the Time of Flight-ERD spectrometer used to measure energy loss of heavy ions through freestanding stopper foils.