SAND99-1909C

Quantitative analysis with heavy ion E-TOF ERD

A. Climent Font [a*], J.C. Banks [b] and B.L. Doyle [b]

[a] *Universidad Autónoma de Madrid, Depto. Fisica Aplicada C-12 and Instituto Nicolas Cabrera, Cantoblanco E-28049 Madrid, Spain,*

[b] *Sandia National Laboratories, Radiation Solid Interactions and Processing, Albuquerque, NM 87185-1056, USA.*

Abstract

Heavy ion TOF ERD combined with energy detection (E-TOF-ERD) is a powerful analytical technique taking advantage of the following facts: the scattering cross section is usually very high (~10^{-21} cm^2/sr) compared to regular He RBS (~10^{-25} cm^2/sr), contrary to what happens with the energy resolution in ordinary surface solid barrier detectors, time resolution is almost independent of the atomic mass of the detected element, and the detection in coincidence of time and energy signals allows for the mass separation of overlapping signals with the same energy (or time of flight). Measurements on several oxides have been performed with the E-TOF-ERD set up at Sandia National Laboratories using an incident beam of 10-15 MeV Au. The information on the composition of the sample is obtained from the time domain spectrum, which is converted to energy domain, and then, using existing software codes, the analysis is performed. During the quantification of the results, we have found problems related to the interaction of the beam with the sample and to the tabulated values of the stopping powers for heavy ions.

Keywords: Heavy ion elastic recoil detection; Energy and time of flight ERD; quantification of E-TOF-ERD spectra.

*Corresponding author. Aurelio Climent-Font. Departamento de Física Aplicada C-XII and Instituto Nicolás Cabrera. Universidad Autónoma de Madrid, Cantoblanco E-28049 Madrid, Spain. Tel.: +34 91 397 5264; fax: +34 91 397 3969; e-mail: acf@uam.es.

DISCLAIMER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

1. Introduction

Elastic recoil detection, ERD, is a very useful technique for the detection of light elements in a heavier matrix [1-4]. Experimental requirements restrict the detected elements to be lighter than the beam element. In order to prevent the incident particles scattered from the matrix to reach the detector a range foil is installed in front of it. H profiles [2] can thus be obtained with He beams. Using heavier beams, like Si ions [5], elements like C, N, and O can also be detected. The right choice of the beam energy and range foil thickness can avoid the possible overlap of signals when analyzing a thin film. As the atomic number of the recoiled particles increases, its energy detection becomes more problematic using the conventional energy detectors because the resolution is progressively degraded [6]. Besides, the energy spread of the particles passing through the range foil is broadened increasing further the degradation of the energy resolution. To circumvent these problems a number of methods have been reported including ΔE-E recoil telescopes [7,8] and energy and time-of-flight ERD recoil telescopes, E-TOF-ERD, [9-12]. The E-TOF-ERD solution is often used because of the good mass resolution of the time of flight measurement.

In this paper we summarize our results on the quantification of E-TOF-ERD spectra using the coincidence time-energy plot to separate overlapping signals of particles having different mass but the same energy or velocity. The time domain spectra, with a superior depth resolution are converted to energy domain spectra, which are analyzed with the help of available computer codes simulating E-ERD spectra. Finally, we discuss a few problems encountered during the quantification of the spectra, and which are related to the interaction incident beam sample and to the tabulated values of the stopping powers for heavy projectiles which seem inappropriate

2. Experimental

The E-TOF ERD set up at Sandia National Laboratories consists of two Zebelman type [13] pick up time units set 1 m apart and a conventional surface barrier detector [14]. The start and stop signals are generated by the electrons released when an ions passes through a thin C foil (about 20 nm thick in our case). This set up is similar to the one used by Thomas et al. [9], although in our case we use the energy signal not only to gate the time signal, but to get as well the energy spectrum allowing for the

mass separation as stated earlier. The time pickup unit further away from the sample gives the start signal and the closer one gives the stop signal through a delay of a few hundred of ns. With that configuration the faster particles appear in the higher time channels. The time of flight detection line is at 30 ° from the incident beam direction, and the sample is oriented with a goniometer its normal forming an angle of 75° with the direction of the incident beam and the direction of the time of flight path. The time resolution measured when recording an spectrum of SiO_2 with a beam of 10 MeV Au is about 1 ns, which is enough to have a resolution of 1 amu.

The energy and time signals are recorded in coincidence with a PC based multiparameter acquisition system (FAST ComTec). If needed, independent energy and time spectra without coincidence are also available. This software provides the plotting support to select from the time-energy plot of the signals detected in coincidence the convenient areas for the mass separation of the signals.

In the results presented here the incident beam was mostly 10 MeV Au+3, although we have also performed experiments with Au at higher energies (15 MeV) and with other beams (I+2 at 6.5 MeV).

3. Results and discussion

2.1. Dose and solid angle determination

The incident Au beam irradiated an area of the sample of typically 2x7 mm^2. Using a beam of heavy ions the production of secondary electrons and recoiled species in the interaction beam sample is very important and it is difficult to keep all the charged particles produced inside the Faraday cage, affecting a proper determination of the irradiation dose and detector solid angle. To overcome this problem we measured the dose and solid angle by measuring the Au implanted in a sample scanning across the implanted area a 1 mm diameter He beam while recording RBS spectra. The amount of Au was deduced using a standard sample of Bi implanted on Si. Fig. 1 shows the profile of the Au implantation.

2.2. Quantification of E-TOF-ERD measurements

Fig. 2 (a) and (b) shows the energy and time spectra of a SiO_2 film grown on a Si wafer. The Si and O have been separated from the time-energy plot. The continuous line in the energy spectra is the simulation generated with the program SIMNRA [15]. Simulating the spectra we systematically found that the Si signal required about 30 % more dose than the O signal in order to have a good fit between the experimental spectrum and the simulation. This could be due to the time detector being more efficient for the Si than for the O. Simulation with TRIM [16] to get the average angle of dispersion in the time pick up foils and a distribution of the dispersed particles as a function of the exit angle, gave similar results for both atomic species. Later experiments on other thin films containing O like Al_2O_3 films grown on Al, gave the same efficiency for Al and O, therefore the observed fact for SiO_2 samples must have an origin related to the sample character. The time domain spectra can be converted to time domain. A simple BASIC program was written to read the time domain spectrum and convert it to energy domain using the relation of the kinetic energy, $E = (1/2)m(L/t)^2$, where m is the mass of the particle, L the fixed length between the two pick up time units and t the time of flight of the particle. Fig. 2 (c) shows the time converted spectra and the simulations generated with SIMNRA. As it happened with the original energy spectra, in the time-converted spectra, the simulation of the Si signal required a higher dose than the O signal. The improved depth resolution of these time-converted spectra is clearly seen when comparing them to the original energy ones.

2.3. Stopping power problems for heavy ions

The stopping power of the moving ion in the matrix of the sample under analysis is of paramount importance to perform quantitative analysis with ion beam techniques. As stated we used the code SIMNRA to simulate the ERD spectra for its quantification. This code provides the choice of two tables of stopping powers, denoted Andersen-Ziegler and Ziegler-Biersack, to fit the spectra[15]. For the case of Au ions in a SiO_2 matrix these values disagree as shows Fig. 3. The disagreement is specially high for 10 MeV, the energy of our incident Au beam. He RBS measurements on samples later measured by E-TOF-ERD showed that the Andersen-Ziegler values gave the same thickness for the films in both techniques, while the Ziegler-Biersack values gave much more thinner values for the film thickness in the E-TOF-ERD technique. Similar results where observed for Si_3N_4 samples.

2.4. Interaction incident beam sample

While performing repeated E-TOF-ERD measurements with 10 MeV Au ions on SiO_2 films grown on Si substrate the oxide film was eroded away rather fast as noticed by the change in shape of the O and Si signals of either the time or energy spectra. The quantification of the etching process, that is the oxide atoms removed per incident Au ion, was performed by taking successive spectra. Two types of measurements were made: ones meant to measure the thickness of the oxide (with lower irradiation dose) $_2$and other ones, with higher irradiation dose, meant to erode the oxide. The thickness of the remaining oxide layer was measured simulating the ERD spectra with the SIMNRA code and from the area of the O signal of either the energy or time spectra separated from the coincidence time-energy plot using the process described above in paragraphs 2.1. and 2.2. Fig. 4 shows a plot of the remaining SiO_2 layer after a given dose of Au $_{O32ions}$. An etching rate of about 200 SiO_2 atoms per incident Au atoms is obtained from the plot. Simulation made with TRIM gave values of about 35 atoms per incident ion. In the plot it can be seen an small amount of oxide resisting to be eroded away. Comparing the spectra of the remaining oxide with E-TOF-ERD spectra of fresh oxide 5 nm thick, it is observed that while for the fresh spectra the O signal either in the energy or time spectra is thin and tall, the same signal for the remaining oxide is wider and more rounded. This may be due to corrugations formed by the bombarding ions which make more difficult a complete removal of the oxygen. Using other beans like 6.5 MeV I (to have the same energy per nucleon than 10 MeV Au), or 15 MeV Au similar effects of fast etch rate have been observed for SiO_2. Other materials like Al_2O_3 on Al or CuO on Cu have not shown such behavior and no appreciable thinning of the oxide was observed after rather long measurements. If we consider that such a fast erosion rate could be due to a coulombic explosion because of a fast accumulation of charge, this hypothesis would explain the unobserved effects on Al_2O_3 or CuO as those films are formed on a conducting metallic substrate which are able of relaxing much faster the accumulated electrical charge than the Si substrate.

Summary

E-TOF-ERD is a technique capable of giving valuable information because of the possibility of mass separation and an improved depth resolution with the detection of time of flight. It is very easy to convert a time domain spectrum to the energy domain and then use the existing simulation computer codes as a help for the quantification of the results, although it would nice and not more difficult that for the energy analysis to write programs to simulate time spectra. Care must de taken, when using heavy ions, to use reliable stopping power values and be cautious about the interaction effects of the beam with the sample. As the heavy ions produce a lot of electrons and recoils from the sample, especial care must be taken for the determination of the irradiation dose.

References

[1] J. L'Ecuyer, C.- Brassard, C. Cardinal, J. Chabbal, L. Deschenes, J.P. Labrie, B. Terrault, J.G. Martel and St.-Jacques, J. Appl. Phys. 47 (1976) 381.

[2] B.L. Doyle and P.S. Peercy, Appl. Phys. Lett. 34 (1979) 811.

[3] J. Tirira, P. Trocellier, J.P Frontier and P. Trouslard, Nucl. Instr. and Meth. B 45 (1990) 203.

[4] Y. Wang, B. Huang, D. Cao, J. Cao, D. Zhu and K. Shen, Nucl. Instr. and Meth. B 84 (1994) 111.

[5] B.L. Doyle and D.K. Brice, Nucl. Instr. and Meth. B 35 (1988) 301.

[6] D.J. O'Connor and TAN Chunyu, Nucl. Instr. and Meth. B 36 (1989) 178.

[7] J.P. Stockert, G. Guillaume, M. Hage-Ali, J.J. Grob, C. Ganter and P. Siffert, Nucl. Instr. and Meth. B 44 (1989) 184.

[8] W. Assmann, Th. Reichelt, T. Eisenhammer, H. Huber, A. Mahr, H. Schellinger and R. Wohlgemuth, Nucl. Instr. and Meth. B 93 (1996) 303.

[9] J.P. Thomas, M. Fallavier and A. Ziani, Nucl. Instr. and Meth. B 15 (1986) 443

[10] H.J. Withlow, G. Possnert and C.S. Petersson, Nucl. Instr. and Meth. B 27 (1987) 448.

[11] J. H. Arps and R.A. Weller, Nucl. Instr. and Meth. B 100 (1995) 331.

[12] W. Hong, S. Hayakawa, K. Maeda, s. Fukuda, M. Yanokura, M. Aratani, K. Kimura, Y. Gohshi and I. Tanihata, Nucl. Instr. and Meth. B 124 (1997) 95.

[13] A.M. Zebelman, W.G. Meyer, K. Halbach, A.M. Poskanzer, R.G. Sextro. G. Gabor and D.A. Landis, Nucl. Instr. and Meth 141 (1977) 439.

[14] J.A. Knapp, J.C. Barbour and B.L. Doyle, J. Vac. Sci. Technol. A10 (1992) 2685.

[15] M. Mayer, SIMNRA User's Guide. IPP-Report 9/113. Max-Plank-Institute für Plasmaphysik. Garching (1997).
[16] Ref TRIM

Figure captions

Figure 1. Profile of the Au implanted in a SiO2 sample measured by E-TOF-ERD for a long time. The profile was obtained by RBS scanning a He beam of about 1 mm diameter across the 2x8 mm2 implanted area.

Figure 2. (a) Separated Si and O energy spectra obtained from the coincidence time-energy plot. In continuous line are the simulated spectra as described in the text. (b) Same signals separated in the time domain. (c) Energy spectra obtained from the time domain spectra. The simulations are shown in continuous line. Note the improved depth resolution of these spectra when compared to the original energy ones.

Figure 3. The two sets of stopping power values for Au in SiO_2 tabulated in the RBS and ERD simulation code SIMNRA. In E-TOF-ERD experiments with 10 MeV Au ions, Andersen- Ziegler values give results in agreement with He RBS measurements.

Figure 4. An etching rate of about 200 SiO_2 atoms per incident Au ion can be deduced from this plot of the measured oxide thickness as a function of the irradiation dose with 10 MeV Au ions.