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Ion beam effects of 26.0 MeV Cu^{7+} ions in thin metallic and insulating films during Heavy Ion ERDA measurements

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ABSTRACT

We report on an investigation carried out to determine effects of the probing beam on the structure of typical metallic and insulating thin films during Elastic Recoil Detection Analysis (ERDA) using a heavy ion beam. Metallic niobium nitride (NbN) and insulating calcium fluoride (CaF_2) thin films (used as test samples) were irradiated by 26.0 MeV $^{63}Cu^{7+}$ ions to fluences of 1.70×10^{14} ions/cm^2 and 2.70×10^{14} ions/cm^2, respectively. The effects of irradiation on the structural properties of the films were studied using Rutherford Backscattering Spectrometry (RBS), X-ray diffraction (XRD) and Atomic Force Microscopy (AFM). RBS results showed a significant (18%) reduction in the thickness of the CaF_2 film due to electronic sputtering compared to only 1% reduction in the NbN film. XRD results showed no significant peak shifts in both films, but rather formation of unidentified peaks in the insulating film. AFM results indicated a substantial decrease in the average surface roughness of the insulating film and only a nominal increase in that of the metallic film. Results of electronic sputtering yield measurements carried out by ERDA are explained in terms of both the Coulomb explosion and the inelastic thermal spike models.

1. Introduction

Thin solid films are used in a wide range of applications in modern technology from decorative, optical and hard protective coatings through to high frequency filters, solar cells and wire sensors [1]. Transition metal nitride thin films for example are found in many applications due to a combination of refractory and metallic properties. Niobium nitride (NbN) for instance is used as a superconductor and in detectors for infrared light [2]. It has attracted much attention due to its interesting physical properties and the associated technological applications, next to those of titanium nitride [3]. As another example, thin film alkali-earth fluorides and multilayer structures that include such layers are known to be prospective materials for optics, photonics and chemical sensors. Calcium fluoride (CaF_2) films in particular have been used for a long time in many optical components due to its exceptional transparency in the ultra violet (UV) as well as in the infra-red (IR) spectral domain [4].

Many advanced thin-film materials such as the examples cited above derive their functional properties from compounds containing light elements. Nuclear analytical techniques have become indispensable in the development of thin film based technologies. Ion beam analysis (IBA) techniques that use ion beams from particle accelerators have a competitive advantage in that they can provide important information about the concentration, distribution and location of light elements in thin film matrices much more readily and directly than non-nuclear spectrometries [5].

Elastic Recoil Detection Analysis (ERDA) is an IBA technique based on the elastic forward recoiling of target atomic nuclei (generally) lighter than the projectile [6]. It is a complementary scattering technique to Rutherford Backscattering Spectrometry (RBS) and is particularly useful for easy depth profiling of hydrogen (H, D, and T) and helium isotopes in surfaces and thin films. The variant of ERDA using projectiles heavier than helium is known as Heavy Ion ERDA. When performed with He^+ projectiles, ERDA is generally non-destructive, but as the incident ion beam particles become heavier, the effect of the beam on the surface layers of the target sample may become significant, depending on the beam energy and fluence, angle of incidence and nature of the target [7–9]. At MeV energies the dominant mode of beam-target energy exchange is electronic stopping (leading to electronic sputtering) [7]. Much of the work done on electronic sputtering in the grazing incidence ERDA configuration has focused on relatively high

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energy (>1.0 MeV/u) incident ions [10–13]. At lower energies ($0.1 \leqslant E \leqslant 0.5$ MeV/u), apart from work by Gordillo et al. [14], not much else has been reported. This latter energy region is where most small accelerator labs involved in heavy ion Time-of-Flight (ToF) ERDA operate [9,15–20]. The Heavy Ion ERDA technique at iThemba LABS Gauteng [19] was set up recently and there are continual studies to optimise its performance subject to existing infrastructural limitations. This study investigates the effects of the probing beam on the structure (i.e. thickness, crystallinity and roughness) of typical metallic and insulating thin films during ERD analysis using 26.0 MeV $^{63}Cu^{7+}$ ion beams.

2. Experimental

Thin films of NbN/Si were grown by reactive magnetron sputtering of a NbN target. The target was sputtered by Ar^+ ions for 15 min at 100 W RF power. Pre-cleaned silicon substrates were placed 50 mm above the target for film deposition. The base pressure of the system was below 7.2×10^{-1} mbar during the sputtering process. CaF_2/Si samples were prepared by electron beam evaporation of granular CaF_2 onto silicon substrate pieces. The base pressure in the evaporation chamber was maintained below 10^{-7} mbar during deposition. Both film types were characterised for thickness, crystallinity and surface roughness before and after Heavy Ion ERD irradiation analysis.

Irradiation of the films was carried out in the ERDA configuration using the 6 MV Tandem accelerator at iThemba LABS Gauteng. The NbN/Si and CaF_2/Si thin films were each irradiated by a beam of 26.0 MeV $^{63}Cu^{7+}$ ions at average beam currents of 13 nA and 16 nA, respectively, corresponding to ion rates of 1.2×10^{10} s^{-1} and 1.4×10^{10} s^{-1}. The projectile beam hit the target samples at a grazing incidence angle of 15° to the sample surface. The ToF-energy detector telescope used for the detection and identification of recoil ions is set at a forward scattering angle of 30° to the initial beam direction [19]. The beam fluence Φ was calculated from the irradiation time (t), beam current (I), and the beam spot area (A) on the target:

$$\Phi = \frac{It}{qeA}, \tag{1}$$

where q is the ionic charge (7) and e is the electron charge (1.602×10^{-19} C).

Pressure in the target chamber during Heavy Ion ERDA measurements was in the order of 10^{-7} mbar. The metallic film was irradiated to a fluence of 1.70×10^{14} ions/cm^2 and the insulating film was irradiated to 2.70×10^{14} ions/cm^2. Structural characterisation of both films was carried out before and after Cu irradiation to ascertain the effect of the probing beam on the films. Rutherford Backscattering (RBS) measurements were carried out at the University of Pretoria's 2 MV Van De Graff accelerator using 1.6 MeV He^+ ions to measure (any) variation in the thickness and composition of the films. The analysing particle beam was collimated to a spot of 1 mm diameter. A ring-shaped electrode in front of the target was kept at a negative potential of 200 V to suppress secondary electrons. Sufficient counting statistics was obtained by collecting an integrated charge of 8 μC. Backscattered particles were measured at 165° by a surface barrier detector telescope with an acceptance angle of 2°. XRD analyses were performed using a Bruker D8 Advance ™ diffractometer to determine possible beam effects on the structure of the films. The diffractometer was operated at a voltage of 40 kV and a current of 40 mA, generating 8 keV K_α X-rays from a copper target. AFM analyses were carried out using the Dimension Icon® Atomic Force Microscope from Bruker™ to measure the surface roughness and morphology of the films.

3. Results and discussion

3.1. Film thickness measurement results

RBS energy spectra of the metallic NbN and the insulating CaF_2 films obtained before and after irradiation are shown in Figs. 1a and 1b, where the horizontal axis represents backscattered beam energy (in channel numbers). The SIMNRA simulation code [21] was used to obtain the thickness and stoichiometry of the films. However, the simulation plots are not included in Figs. 1a and 1b to avoid obscuring the comparison between the pristine and irradiated films. Both films were found to be stoichiometric, before and after irradiation. The arrows in both figures indicate the surface energy positions of the different atomic species detected in the films. In Fig. 1a, what appears as a nitrogen surface peak is readily fitted by assuming presence of a native SiO_2 layer on the Si substrate. The thickness of the sandwiched oxide layer was estimated to be about 150×10^{15} at/cm^2. Iron contamination, possibly a residual impurity in the RF sputtering chamber, was detected in the NbN film. The uncertainty in the measured thickness values is a convolution several contributions. The main contributor is the uncertainty in the SRIM stopping force for He ions traversing the films – 3.5% [22]. Other (minor) factors include the RBS detector energy resolution (quoted at 20 keV at 5.46 MeV), 0.5% in the beam energy spread and 0.21% uncertainty in the SIMNRA code [23]. All these factors add up to an effective 3.6% uncertainty in the film thickness.

The thickness values obtained from the SIMNRA simulations are summarised in Table 1. There was a small, rather insignificant (<1%) decrease in the thickness of the metallic film after irradiation, while a significant (18%) reduction in thickness was observed in the insulating film. Similar behaviour in other types of insulating films has also been observed by other workers [7–9].

3.2. Phase identification analysis

For phase identification XRD data analysis was done with the proprietary Bruker EVA™ search and match software. The identified phases in the NbN and CaF_2 films are displayed in Figs. 2a and 2b. The XRD spectrum of the metallic film before ion irradiation shows Bragg reflections at 2θ equal to 35°, 39°, 58° and 71° corresponding respectively to (111), (200), (220) and (311) reflections of the NbN face centered phase. After irradiation the Bragg reflections are at the same 2θ values indicating no peak shift

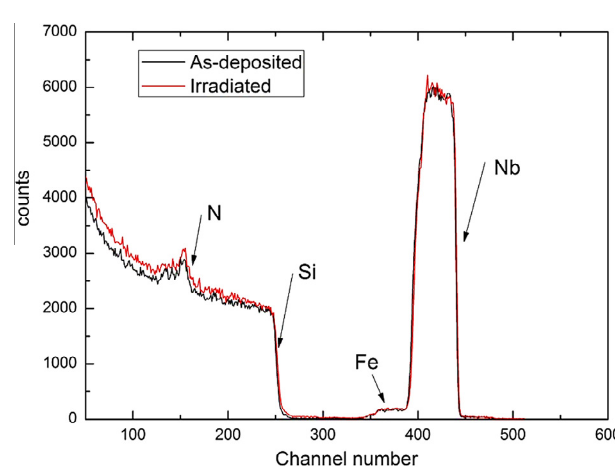

Fig. 1a. RBS spectrum of the as-deposited (black) and Cu-irradiated (red) NbN/Si layer. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)