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Study of deuteron elastic scattering on ^{nat}N , by implementing the EBS technique

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Abstract In the present work, the elastic scattering of deuterons on ^{nat}N has been studied in the energy range of $E_{d,\text{lab}}=1000\text{--}2200$ keV, in steps of 10 keV, at six backscattering detection angles (120° , 130° , 140° , 150° , 160° and 170°), suitable for analytical purposes. The target used for the measurements was a self-supported Si_3N_4 thin foil, with an ultra-thin Au layer evaporated on top for normalization purposes. The measurements were performed using the 5.5 MV TN11 HV Tandem Accelerator of N.C.S.R. “Demokritos”, Athens, Greece and a high-precision goniometer. The effect of the various nuclear reaction mechanisms in low-energy deuteron elastic scattering are discussed and the obtained differential cross-section datasets are compared to already existing ones in literature.

Keywords Elastic Scattering, EBS, ^{nat}N , Differential cross sections

INTRODUCTION

Nitrogen is the seventh most abundant element in the solar system, comprising in molecular form, ca. 78% of the earth’s atmosphere and is composed of two natural isotopes, ^{14}N (99.634%) and ^{15}N (0.366%). It has numerous applications in metallurgy, as well as in semiconductor- and insulator technology. More specifically, the implantation of nitrogen into metals, e.g. steels, titanium and titanium alloys, increases their hardness and wear resistance, an important feature for cutting tools. Additionally, nitrogen is a common dopant for the creation of n-type semiconductors, and it is also used as an additive to the argon atmosphere during the crystal growing process, while a nitrogen-containing passivation layer prevents the increase and migration of displacements during the annealing process.

Despite the fact that the accurate quantitative determination and depth profiling of nitrogen in various targets constitutes a major challenge for all Ion Beam Analysis (IBA) techniques, it has been the subject of extensive, pioneer works in the past (e.g. [1, 2]). The major difficulty is that nitrogen usually coexists in various complex matrices along with several other low- and medium-Z elements. Nevertheless, there is a certain lack of coherent experimental differential cross-section datasets for nitrogen, especially of those concerning the deuteron elastic backscattering spectroscopy (d-EBS) technique.

In order to address this problem the elastic scattering of deuterons on ^{nat}N has been

studied for deuteron beam energies and backscattering detection angles, suitable for analytical purposes.

EXPERIMENTAL DETAILS

The experiment proceeded in several distinct phases, with the d-EBS experiments having been performed using the deuteron beam of the 5.5 MV TN11 HV Tandem Accelerator of N.C.S.R. “Demokritos”, Athens, Greece. The deuterons, accelerated to $E_{d,lab}=1000\text{--}2200$ keV, were directed to a large-size, cylindrical scattering chamber (radius: 40 cm) equipped with a high-precision goniometer (0.1°). The final ion energy of the deuteron beam was determined by Nuclear Magnetic Resonance (NMR) with an estimated ripple of $\sim 1.5\%$, as verified at the beginning of the experiment – using protons – by the reaction rate of the 991.89 keV resonance of the $^{27}\text{Al}(p,\gamma)$ reaction, using a 18% relative efficiency HPGe detector. Since non-linear deviations of the magnet have not been observed in the past, the determined energy offset (~ 3.3 keV) and ripple (~ 3 keV) were taken as constant for the limited deuteron beam energy range studied in the present work. Differential cross-sections were obtained using a constant beam energy step of 10 keV, since no fine structure was expected in the measured excitation functions.

Six Si surface barrier detectors (thickness: 500 μm ; set at 10° intervals, between 120° and 170°) consisted the detection system, along with the standard corresponding spectroscopy electronics, as the target was placed at a distance of $\sim 8\text{--}12$ cm from the detectors. The beam spot size was limited to $1\times 1\text{ mm}^2$, while the current on the target did not exceed ~ 100 nA during all measurements, in order to avoid pileup effects and overheating of the (thin) target. The vacuum in the chamber was kept constant and as low as $\sim 1\times 10^{-6}$ Torr.

A new version of SIMNRA (v. 6.94) [6] was used in order to calculate the mean deuteron beam energy at half the target’s thickness, following the determination of the target composition via the analysis of proton spectra taken at $E_{p,lab}=980, 1200$ and 1300 keV exactly for this purpose, using the same experimental setup. The analysis of the EBS spectra was accomplished taking into account a very small energy step for the incoming and outgoing protons, the exact detector geometry, the effect of multiple scattering, the beam ripple, ZBL stopping power data along with recent corrections for silicon [7-8], and Chu and Yang’s straggling model as implemented in the code.

The target used for the measurements was an ultra-thin, self-supported, high-purity silicon nitride membrane, having a nominal thickness of 75 nm (Silson Ltd) and an active area of $5\times 5\text{ mm}^2$. As mentioned above, an ultra-thin Au layer ($\sim 14\times 10^{15}$ at/cm²) was evaporated on top of the membrane for normalization purposes. The main problem of these silicon nitride targets is related to the variation of the Si:N ratio, which strongly affects the membrane fragility.

RESULTS AND DISCUSSION

The differential cross sections were determined using the corresponding formula of the relative measurement technique [9]:

$$\left(\frac{d\sigma}{d\Omega}\right)_{E,\theta}^N = \left(\frac{d\sigma}{d\Omega}\right)_{E',\theta}^{Au,Ruth} \times \frac{Y_N}{Y_{Au}} \times \frac{N_{Au}}{N_N}$$

where θ corresponds to the scattering angle, E and E' represent the energies at the half of the target's thickness and at the surface of the target (following the accelerator energy calibration) respectively, Y_N and Y_{Au} are the integrated yields as obtained from the experimental spectra and N_{Au}/N_N is the ratio of the total number of Au versus ^{nat}N nuclei present in the target.

The cross-section of deuteron elastic scattering from gold, $\left(\frac{d\sigma}{d\Omega}\right)_{E',\theta}^{Au,Ruth}$, was calculated according to the Rutherford formula, along with screening corrections over the whole energy range under study ($E_{d,lab}=1000-2200$ keV).

Although d-induced EBS spectra are generally relatively rich, there was no peak overlap, or significant induced background contribution under the nitrogen and gold elastic peaks for the whole energy range under study. Moreover, the carbon and oxygen unavoidable parasitic contributions were clearly separated from the nitrogen peak. The statistical error in Y_{Au} was always kept below 1%, while the corresponding one in Y_N did not exceed 3.5% in the least favorable case.

The accurate determination of the N_{Au}/N_{natN} ratio is probably the most crucial factor for the determination of the differential cross-section values using the relative technique. Thus, since deuteron elastic scattering from nitrogen was *ab initio* expected to present strong deviations from the Rutherford formula over the whole deuteron beam energy range covered in the present work, measurements with protons as projectiles, at three different beam energies (namely at $E_{p,lab}=980, 1200$ and 1300 keV), were also performed. Unlike deuteron elastic scattering, proton elastic scattering on nitrogen has been well evaluated and benchmarked [10]. Additionally, in order to maximize accuracy, the proton beam energies were taken far from existing sharp, Breit-Wigner type resonances. For the resulting spectral analysis using SIMNRA, the $Q \times \Omega$ product was determined from the non-Rutherford evaluated data for proton elastic scattering on ^{nat}Si , based on [11], using the online calculator SigmaCalc 2.0, (from <http://sigmacalc.iate.obninsk.ru/>). Following this procedure, the total Au and N thickness (in atoms/cm²) was determined for every proton beam/scattering angle combination using the same experimental setup as in the differential cross-section measurements with the deuteron beam, by slightly varying the target composition in each case. The average value of the N_{Au}/N_{natN} ratio was thus determined to be 0.0423 ± 0.0011 , with a relative statistical error of $\sim 2.6\%$. This error does not include any systematic uncertainties, originating mainly from the accuracy of implemented stopping power compilations, lateral inhomogeneities in the target composition and carbon buildup effects, which –in any case– did not exceed 4-5% (in total).

Following this simulation, the average obtained target composition was used for the determination of E , at each beam energy step, as follows: $E = E' - \Delta E_{Au} - \Delta E_{N/2}$, where E' corresponds to the corrected accelerator energy, ΔE_{Au} corresponds to the energy loss in the ultra-thin surface gold layer and $\Delta E_{N/2}$ to the energy loss taking into account half of the silicon nitride target thickness. These E values were subsequently used as the appropriate x -values in the corresponding differential cross-section figures and for the accurate determination of the $\left(\frac{d\sigma}{d\Omega}\right)_{E,\theta}^{N,Ruth}$ values, including screening effects, in cases where the results are presented in a 'ratio to Rutherford' form.

The differential cross-section values for the deuteron elastic scattering on nitrogen, $^{nat}N(d,d_0)$ measured for the six backscattering angles are presented in the following graphs (Fig 1a-f) and compared with results from previous measurements according to the closest experimental angle under study. The combined experimental statistical uncertainty did not exceed 5.3% in all cases.

Fig. 1a-f. The differential cross-section for the deuteron elastic scattering from ^{nat}N

The effect of the resonant mechanism in the obtained results can be better assessed if one examines the ratio of the obtained values for nitrogen to the ones calculated using Rutherford's formula. These ratios, for all scattering angles studied in the present work are presented in Fig 2a-f. It is evident that despite the fact that the deuteron elastic cross-section

values remain lower than the Rutherford ones over the whole energy range studied, there is a strong, increasing trend for deuteron beam lab energies above ~ 1800 – 1900 keV and for steep backscattering angles. This trend is progressively increasing and could be attributed to the multiple overlapping levels of the compound nucleus ^{16}O . On the other hand, it is difficult to estimate the direct reaction mechanism contribution, which is also expected to increase with increasing deuteron beam energy, but early results for the elastic channel [4] did not render it as overall important.

Fig. 2a-f. The differential cross-section ratio to Rutherford for the deuteron elastic scattering from ^{nat}N

CONCLUSIONS

In the present work differential cross-section values for deuteron elastic scattering from ^{nat}N were determined in the energy range $E_{d,lab} = 1000$ – 2200 keV in steps of 10 keV, for six backscattering angles, namely at 120° , 130° , 140° , 150° , 160° , 170° , suitable for Elastic Backscattering Spectroscopy. Using the relative technique and a solid-state target, a coherent set of differential cross-section measurements has been obtained, avoiding the additional experimental uncertainties caused by variations in the target thickness and the $Q \times \Omega$ product.

The obtained values showed large deviations from the Rutherford formula, which can be mainly attributed to the broad, overlapping resonances of the compound nucleus $^{16}\text{O}^*$. The datasets obtained in the present work are expected to facilitate future analytical combined EBS/NRA nitrogen depth profiling studies since the results will soon be available to the scientific community at the IBANDL website (<https://www-nds.iaea.org/exfor/ibandl.htm>), in both tabulated and graphical forms.

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