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Cross-section for $^{14}N(\alpha, p_0)^{17}O$ reaction in the energy range 3.2–4.0 MeV

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Abstract

The cross-section for the $^{14}N(\alpha, p_0)^{17}O$ nuclear reaction was measured in the beam energy range between 3.2 and 4.0 MeV, at a laboratory scattering angle of 135° using a thin TiN film (95 nm) deposited on Si substrate. The accurate stoichiometry as well as the thickness of the TiN film and other potential impurities were characterized by ERD–TOF with a 40 MeV Cu^{8+} beam and RBS with an 800 keV He^{+} beam. The cross-section was evaluated at maximum intervals of 15 keV, with smaller intervals of less than 10 keV in the neighborhood of the resonance, from the simultaneously measured nuclear reaction spectra and Rutherford backscattering spectra. The total uncertainty of the cross-section is estimated to be less than 7%. As an application, the evaluated cross-section was used to simulate the NRA spectra obtained from a $GaAs_{1-x}N_x$ sample with different beam energies. The nitrogen concentration results are in good agreement with the secondary ion mass spectrometry measurements.

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Keywords: Cross-section; Nuclear reaction; Nitrogen; Helium

1. Introduction

Among the several nuclear reactions that can be used to characterize nitrogen, such as $^{14}N(d,\alpha)^{12}C$, $^{14}N(d,p)^{15}N$, $^{14}N(\alpha, p_0)^{17}O$ and $^{14}N(\alpha,\gamma)^{18}F$ [1,2], the $^{14}N(\alpha, p_0)^{17}O$ endothermic resonance nuclear reaction stands out to be a better choice in most of ion beam laboratories because other nuclear reactions activated by probing deuterium beam induce unwarranted long-term radiation background in the setup. In the past, Doyle et al. applied $^{14}N(\alpha, p_0)^{17}O$ reaction on the ^{14}N analysis using microbeam [3] and later, Lin et al. measured the excitation curve at 135° from 3.4 to 4.0 MeV which showed a resonance at beam energy around 3.7 MeV [4]. However no absolute cross-sections were presented. More recently, Giorginis et al. measured the cross-section of $^{14}N(\alpha, p_0)^{17}O$ nuclear reaction at 135° with the beam energy in the range 4.0–5.0 MeV [5] and observed three resonance peaks with the most pronounced one being excited at 4.44 MeV. Our preliminary measurements of the cross-sections in the energy range 3.2–4.0 MeV revealed that the maximum of the cross-section at the resonance around 3.7 MeV is comparable to the peak value at 4.44 MeV mentioned earlier. Lower beam energy accounts for a better depth resolution and less energy spread due to the thinner stopping foil needed to stop the scattered ions. In addition, at lower beam energy, the contributions from other possible beam-induced reactions are expected to be lower.

In this paper, we report and discuss the cross-sections measurements for the $^{14}N(\alpha, p_0)^{17}O$ nuclear reaction at a laboratory scattering angle $\theta_{lab} = 135°$ in the α particle energy range 3.2–4.0 MeV. An example using the evaluated cross-sections to characterize the $GaAs_{1-x}N_x$ epitaxial layer is presented at the end of the paper. The results of

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the simulation on NRA spectra are in good agreement with SIMS measurements.

2. Experimental

A commercial TiN thin film (~95 nm) deposited on Si substrate was selected for the measurement of cross-section variation as a function of incident beam energy. In order to obtain the accurate information of the ratio of area densities of Ti and N as well as the thickness of the film, ERD–TOF with 40 MeV Cu^{8+} beams (carried out on a 6 MV tandem accelerator [6]) and RBS with 800 keV He^{+} respectively were carried out. During the RBS measurement, the samples were tilted 7° to avoid the channeling effect in the Si substrate, which was used for normalization in the simulation. In ERD–TOF measurement, the recoil angle was 30° while the angle between the beam and the sample surface was 15°.

The cross-section measurements were carried out in a RBS chamber equipped with a motor controlled three axes goniometer and the He^{2+} beam was obtained from a 1.7 MV Tandetron accelerator at Université de Montréal, in the energy range 3.2–4.0 MeV. The beam energy was calibrated using the resonance in the $^{19}F(p,\alpha\gamma)^{16}O$ nuclear reaction at 872.1 keV, the elastic backscattering resonances $^{16}O(\alpha,\alpha)^{16}O$ at 3363 and 3877 keV, and $^{12}C(\alpha,\alpha)^{12}C$ at 4265 keV. The uncertainty was within ±4 keV in the experimental energy range. A well focused beam of 1 mm in diameter was normally incident on the sample. RBS and NRA spectra were simultaneously detected by two standard passivated implanted planar silicon (PIPS) detectors with ~15 keV energy resolution that were located at ±135° with respect to the direction of incident beam respectively. In order to reduce the uncertainty on the detection angle, the perpendicular angle between beam direction and the sample surface was corrected by aligning the beam along ⟨1 0 0⟩ axis of a piece of (1 0 0) oriented Si single crystal sample through channeling method. Then the detector was aligned with outgoing particle from ⟨1 1 0⟩ axis to define the angle between the detector and the beam to be 45°. The uncertainty of the angle is less than 1°. The NRA detector was mounted at a distance closer to the sample in order to increase the solid angle i.e. improve the detection efficiency. Five or six layers of mylar foil with a total thickness 12.7 or 15.2 μm were placed in front of the NRA detector to stop the α particles scattered from target. In order to determine the ratio of solid angles between the NRA and RBS detector, the RBS spectra from a 25 nm thick Pt film on Si substrate were simultaneously collected by the two detectors and the ratio of solid angle was deduced by comparing the Pt peak areas from the two spectra.

A +300 V bias voltage was applied on the sample to suppress secondary electrons emission. The cross-sections were measured at maximum intervals of 15 keV, with smaller intervals of less than 10 keV used in the neighborhood of resonances. During the data acquisition, the beam current was maintained at ~20 nA while 50 μC of charges were accumulated for each energy step to achieve sufficient statistics. In order to avoid any possible carbon contamination at the sample surface, the beam spot was moved to a fresh target point every three energy steps.

3. Data analysis

The method we used to evaluate the cross-section from the experimental data is based on the comparison of integrated proton peaks in NRA spectra and scattered peaks from Ti in RBS spectra. They are expressed in the following formula:

$$Y_{\mathrm{NRA}} = Q\Omega_{\mathrm{NRA}}C_{\mathrm{N}}\sigma_{\mathrm{NRA}}\left(E_0 - \frac{\Delta E}{2}\right), \tag{1}$$

$$Y_{\mathrm{RBS}} = Q\Omega_{\mathrm{RBS}}C_{\mathrm{Ti}}\sigma_{\mathrm{RBS}}\left(E_0 - \frac{\Delta E}{2}\right), \tag{2}$$

where Y_{NRA} and Y_{RBS} are the integrated yields of proton from $^{14}N(\alpha,p_0)^{17}O$ reaction in NRA spectra and scattered helium from Ti in RBS spectra, respectively. Q is the number of He^{2+} ions bombarding the sample. Ω_{NRA} and Ω_{RBS} are the solid angles of NRA detector and RBS detector, respectively. C_{N} and C_{Ti} are the atomic area densities of N and Ti in the film, respectively. ΔE is the beam energy loss in the layer, $\sigma_{\mathrm{NRA}}(E_0 - \Delta E/2)$ and $\sigma_{\mathrm{RBS}}(E_0 - \Delta E/2)$ are the differential $^{14}N(\alpha,p_0)^{17}O$ NRA and RBS (for Ti) cross-sections, respectively. $(E_0 - \Delta E/2)$ is the effective energy, that is, the average energy at which the reaction occurs in the layer.

From Eqs. (1) and (2), the NRA cross-section is expressed as

$$\sigma_{\mathrm{NRA}}\left(E_0 - \frac{\Delta E}{2}\right) = \frac{Y_{\mathrm{NRA}}\Omega_{\mathrm{RBS}}C_{\mathrm{Ti}}\sigma_{\mathrm{RBS}}\left(E_0 - \frac{\Delta E}{2}\right)}{Y_{\mathrm{RBS}}\Omega_{\mathrm{NRA}}C_{\mathrm{N}}}. \tag{3}$$

The uncertainty of the charge collected is eliminated by using this method and only a properly established ratio of the solid angles and sample stoichiometry are necessary.

4. Results and application

Fig. 1 shows the atomic ratio of Ti and average N in the film measured by ERD–TOF using 40 MeV Cu^{8+} beam. The film is homogeneous and $C_{\mathrm{Ti}}/C_{\mathrm{N}} = 1.00 \pm 0.05$. The thickness of the film is $(960 \pm 10) \times 10^{15}$ at./cm^2. A few percent of O were detected at the surface and at the interface between the TiN film and Si substrate by ERD–TOF. However, the error on the stopping power due to the O presence at surface is less than 1%. The stoichiometry and the thickness of the film were confirmed by RBS measurement using 800 keV He^{+} beam. There are no detectable impurities from RBS spectrum.

The Ti peak is well separated from Si and N signals in the typical RBS spectrum. Therefore, an accurate value of the integrated yields for the Ti peak can be obtained. On the other hand, by carefully choosing the proper

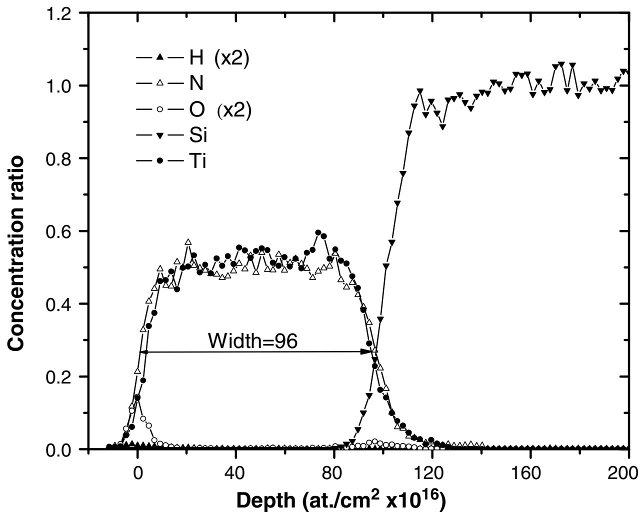

Fig. 1. Depth profile of the sample measured by ERD–TOF using 40 MeV Cu^{8+} beams, the element concentrations were normalized by Si signal.

thickness of stopping foil, a background free proton peak from the $^{14}N(\alpha, p_0)^{17}O$ nuclear reaction can also be obtained. However, with the increase of beam energy, a large yield of protons from the sharp resonance in the $^{28}Si(\alpha, p)^{31}P$ nuclear reaction at 3870 keV appeared in the spectrum. Fortunately, the proton peaks from $^{28}Si(\alpha, p)^{31}P$ and $^{14}N(\alpha, p_0)^{17}O$ are well separated.

The energy loss in the TiN film is estimated to be 37–33 keV for 3.2–4.0 MeV He^{2+} beam energy according to SRIM [7]. The stopping power variation over the whole target thickness was constant within the stopping power uncertainty (1%). The corresponding Rutherford backscattering cross-section, which can be calculated precisely, is valid and smooth in this energy range. For each energy step, Y_{NRA} and Y_{RBS} were obtained by integrating the proton peak from the $^{14}N(\alpha, p_0)^{17}O$ reaction (NRA spectra) and the helium peak backscattered from Ti (RBS spectra), respectively. Taking into account that the ratio of solid angles measured on the thin Pt film: $\Omega_{\mathrm{NRA}}/\Omega_{\mathrm{RBS}} = 2.16$, the corresponding cross-sections can be evaluated using Eq. (3). Results are reported in Fig. 2. The maximum of the cross-section is excited at an energy of 3692 keV. This cross-section value is comparable to the resonance excited at 4440 keV that was published in [5].

Eq. (1) is valid only when the cross-section varies slowly with the energy. From Fig. 2, the FWHM of the resonance peak is about 75 keV, which is larger than the beam energy loss in the layer, although the latter is not negligible in comparison. The result is an apparent broadening of the resonance. To account for this effect, a Lorentzian function convoluted by a boxcar average over $\Delta E/2 = \pm 16.5$ keV was fitted to the experimental data. The empty circles in Fig. 2 show the unconvoluted Lorentzian, which is a more reliable estimate of the cross-section value near the peak. Although the correction on the peak amplitude is 13%, the area in the energy range between 3.5 and 3.8 MeV is affected only by a factor of 2%.

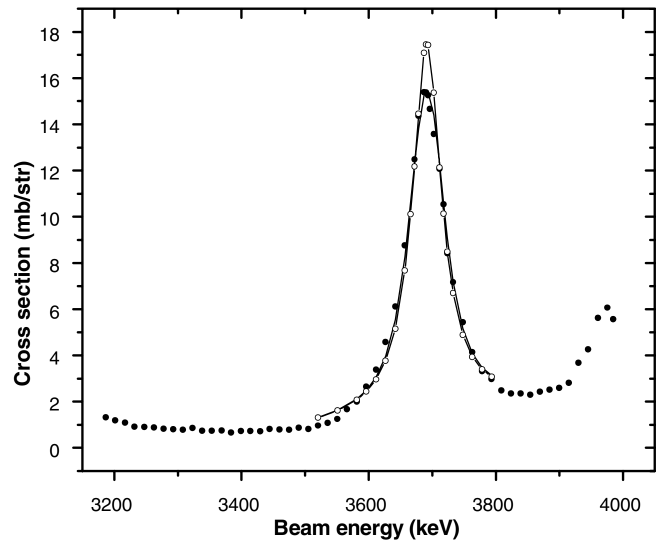

Fig. 2. Energy versus nuclear reaction cross-section in the energy range 3200–4000 keV at a backscattering angle of 135°. Solid circle: experimental data; line: convoluted Lorentzian; empty circle: unconvoluted Lorentzian.

In the error analysis, the contribution from counting statistic was relatively small due to the high N concentration in the target and the relatively high NRA detection efficiency (due to a larger solid angle). The estimated statistical error in Y_{RBS} is ~0.3% while in Y_{NRA} it is better than 3% in the flat region of excitation curve and only 0.7% at the peak of resonance. The error in the experimentally evaluated $\Omega_{\mathrm{RBS}}/\Omega_{\mathrm{NRA}}$ is also determined from counting statistics and is about 0.3%. The uncertainty on the actual detector angles would not account for more than 0.5%. In the ERD–TOF experiments the uncertainty in $C_{\mathrm{Ti}}/C_{\mathrm{N}}$ is ~5%. Therefore the uncertainty in the evaluated cross-section is less than 7%.

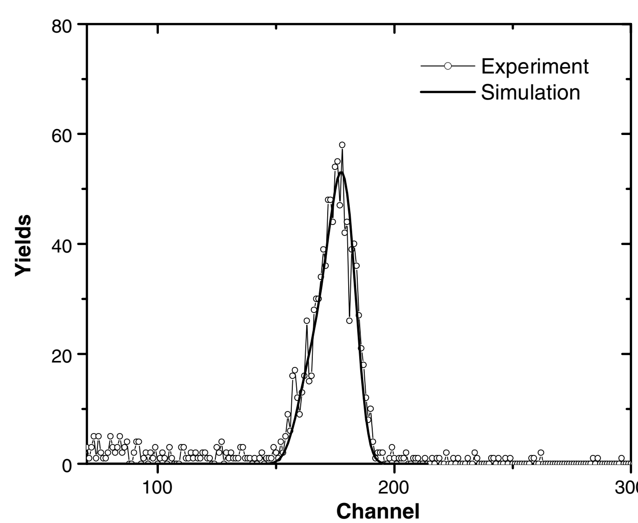

Fig. 3. NRA spectrum from 320 nm thick $GaAs_{1-x}N_x$ film grown on GaAs wafer obtained using 3707 keV He^{2+} beam along with a SIMNRA simulation using the evaluated cross-sections.

In order to check the validity of the evaluated cross-section, a 3707 keV He^{2+} beam was applied for exciting the nuclear reaction on a homogenous $GaAs_{1-x}N_x$ film grown on GaAs wafer by MOVPE [8,9]. The sample was tilted 7° to avoid the channeling effect. NRA and RBS spectra were simultaneously obtained with the same geometry as for the cross-section measurements described above. For comparison, the same sample was also characterized by SIMS technique. SIMS spectrum indicates a thickness of 324 ± 10 nm and the following composition: $GaAs_{0.976\pm0.002}N_{0.024\pm0.002}$. Fig. 3 shows the NRA spectrum as well as the simulation using the SIMNRA program [10]. From the simulation, a thickness of $(1.45 \pm 0.02) \times 10^{18}$ at./cm^2 and a film composition of $GaAs_{0.975\pm0.002}$-$N_{0.025\pm0.002}$ were obtained. Assuming the density of $GaAs_{1-x}N_x$ is the same as GaAs (4.43×10^{22} at./cm^3) due to only a few percent N incorporated in the film, the NRA results are in excellent agreement with the SIMS results. The experiment was repeated at several incident beam energies near the resonance. All the simulation results show the same value within the experimental uncertainty.

5. Conclusions

The cross-section of the $^{14}N(\alpha, p_0)^{17}O$ endothermic nuclear reaction for He beam energy 3.2–4.0 MeV has been evaluated at laboratory scattering angle of 135°. The error of the measurement was estimated to be 7%. As an application, the cross-section was used to simulate the NRA spectra obtained from $GaAs_{1-x}N_x$ sample with different beam energies near the resonance. The simulation results were in good agreement with SIMS measurements. The numerical differential cross-sections will be available soon in Ion Beam Analysis Nuclear Data Library at IAEA web site (http://amdu1.iaea.org/ibandl/).

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