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Angular distribution and recoil effect for 1 MeV Au^+ ions through a Si_3N_4 thin foil

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ABSTRACT

The Stopping and Range of Ions in Matter (SRIM) code has been widely used to predict nuclear stopping power and angular distribution of ion–solid collisions. However, experimental validation of the predictions is insufficient for slow heavy ions in nonmetallic compounds. In this work, time-of-flight secondary ion mass spectrometry (ToF-SIMS) is applied to determine the angular distribution of 1 MeV Au ions after penetrating a Si_3N_4 foil with a thickness of ~100 nm. The exiting Au ions are collected by a Si wafer located ~14 mm behind the Si_3N_4 foil, and the resulting 2-dimensional distribution of Au ions on the Si wafer is measured by ToF-SIMS. The SRIM-predicted angular distribution of Au ions through the Si_3N_4 thin foil is compared with the measured results, indicating that SRIM slightly overestimates the nuclear stopping power by up to 10%. In addition, thickness reduction of the suspended Si_3N_4 foils induced by 1 MeV Au ion irradiation is observed with an average loss rate of ~107 atoms/ion.

1. Introduction

The energy loss of ions penetrating through a target is generally described by two mechanisms, electronic energy loss and nuclear energy loss. Electronic energy loss is attributed to inelastic collisions with target electrons, dominates in the high energy regime, and has little effect on the direction of ions. Nuclear energy loss is attributed to elastic collisions with target atoms, dominates in the low energy regime, and is the primary mechanism for deflection or scattering of ions. Stopping power is defined as the energy loss per unit path length.

In the past decade, experimental data [1–7] have shown that the electronic stopping powers predicted by the Stopping and Range of Ions in Matter (SRIM), a widely used code, are overestimated for very heavy ions (e.g. Au) in low energy regime (e.g., <15 MeV) for targets containing light elements. A transmission setup (Fig. 1) has been developed to directly measure the electronic stopping power [4,8,9]. The energy of ions before and after penetrating a thin foil is measured to determine the energy loss. Electronic stopping power can be determined directly by this method for light ions (H, He) and medium heavy ions (C, Si and etc.). For very heavy ions [4,7] however, nuclear energy loss is not negligible, and must be subtracted from the measured values in order to determine the correct electronic stopping power. In addition, due to the restriction of the collimator placed in front of the Si detector (as shown in Fig. 1), only part of the ions within a certain scattering angle can be detected. In order to accurately determine the electronic stopping power, reliable predictions of scattering angle and nuclear stopping power are critically important. Furthermore, due to the thickness limit of the stopping foil, in the lower energy regime (e.g., <5 MeV), where the electronic stopping powers predicted by SRIM have been shown to have larger errors, the current transmission setup is not able to directly measure the stopping power. As an alternative approach, by measuring the depth distributions of implanted ions with different energies, total stopping power (nuclear plus electronic stopping power) can be derived. With accurate information of nuclear stopping power, the electronic stopping power can be reasonably estimated [4].

The SRIM code has been widely used to predict the scattering process and the nuclear stopping power for several decades [10,11]. However, the predicted nuclear stopping powers of slow heavy ions may be overestimated due to the assumption of neutral

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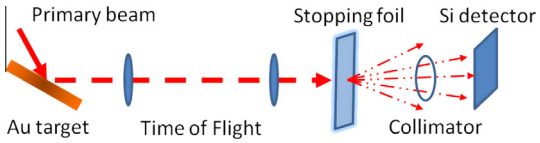

Fig. 1. A transmission setup for electronic stopping power measurement. The primary ion beam is scattered by the Au target. The energy loss of ions is measured by a ToF telescope and a Si detector before and after penetrating the stopping foil. There is a collimator in front of the Si detector that restricts the solid angle of the scattered ions to be detected.

atoms [12]. Direct experimental validation of SRIM predicted nuclear stopping power and scattering angles, especially for very heavy ions in low energy regime, is insufficient. Recently, the reliability of SRIM predicted nuclear stopping power for slow heavy ions has been questioned by Paul [12], which makes direct measurement of angular scattering distributions critically important.

Historically, position-sensitive detectors were used to measure the angular distribution of light ions and medium heavy ions [13]. However, the conventional detectors (i.e. Si detector) are problematic for measuring very heavy ions [14–16]. A time-of-flight spectrometer that could be continuously rotated was used to measure the angular distribution of Bi ions with 0.8 Bohr velocity though a C foil [17]. Unfortunately, this is not a standard instrument for most ion beam laboratories due to special facility requirements.

In this work, a new method is developed to measure the angular distribution of 1 MeV Au ions through a Si_3N_4 thin foil using a time-of-flight secondary ion mass spectrometry (ToF-SIMS). In addition, the thickness of the suspended Si_3N_4 films under ion irradiation is measured as a function of ion fluence by ToF-SIMS, focused ion beam/scanning electron microscope (FIB/SEM) and transmission electron microscope (TEM).

2. Experiments

The angular distribution measurements mainly consist of two parts, ion irradiation and ToF-SIMS measurement. The setup of ion irradiation is shown in Fig. 2. Low stress LPCVD (Low Pressure Chemical Vapor Deposition) Si_3N_4 foils were deposited on a silicon substrate by Norcada Inc (Edmonton, AB, Canada). The foils range from 95 to 103 nm in thickness, and the actual thickness of each foil was measured independently by ToF-SIMS, SEM and TEM. The density of the foils was provided by the manufacturer as 3.1 g cm^{-3}, which has been confirmed by areal density measurement with Rutherford backscattering spectrometry (RBS) in combination with the SEM and TEM results. The Si_3N_4 foils were grown on a Si substrate (5 × 5 mm, 200 μm thick) that is thick enough to block the incoming beam. A window in the central area of the silicon substrate, 0.25 × 0.25 mm, was created by etching [18] and is taken as the beam spot size. The Si_3N_4/Si window was mounted on the front of a metal frame with a thickness of 14.2 mm, and a Si wafer was mounted at the back of this frame. Ion irradiations were performed using the 3.0 MV tandem accelerator at the Environmental Molecular Sciences Laboratory (EMSL) at the Pacific Northwest National Laboratory (PNNL). The Au beam covered the whole 0.25 mm window region, and the transmitted ions were collected by the Si wafer.

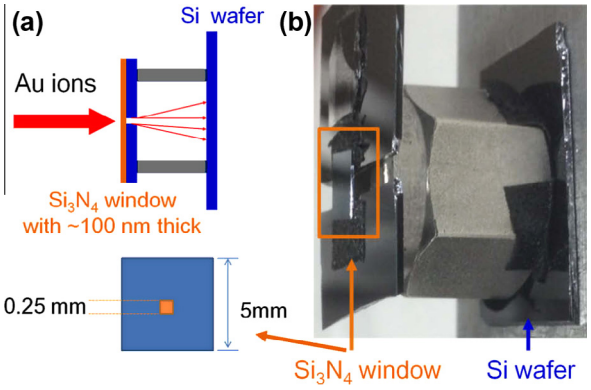

Fig. 2. A schematic illustration (a) and photo (b) of the setup to collect scattered Au ions passing through a Si_3N_4 thin foil. The penetrated Au ions are collected in a Si wafer located at the rear of the metal frame. The suspended Si_3N_4 foil (0.25 mm × 0.25 mm) at the center of the Si wafer is defined as the "window region" and the rest of Si_3N_4 thin film is defined as "supported region" in this work.

The Si wafers implanted with Au ions were subsequently analyzed by ToF-SIMS. Dual-beam mode was used for depth profiling, with 25 keV Bi^+ as the analysis beam and 2 keV Cs^+ as the sputtering beam. As shown in Fig. 3, depth profiling was determined across the Si wafer with steps of 0.5 mm. The size of each crater was 0.3 × 0.3 mm. At each position, the Au depth profile was integrated to calculate the implanted fluence and was normalized to the $^{30}Si^-$ signal in order to minimize the effects from current fluctuations. In order to locate the center of beam, three line scans were performed on each sample: the first scan was performed from a rough center of the wafer; the second scan started from the Au peak position of the first scan along a direction perpendicular to the first scan; and the third scan started from the Au peak position of the second scan along a direction perpendicular to the second scan (parallel to the first scan). Following such approach, the distributions from the second and the third scan are ensured along the diameters of the Au distribution.

In order to evaluate the thickness change of the foils under ion irradiation, ToF-SIMS depth profiling was performed on both the window (the suspended foil without the Si substrate: irradiated area) and the supported region (the film supported by the Si substrate: virgin area) of each sample under different irradiation fluences. The sputtering rate was determined by the crater depth measured by a Dektak 150 profilometer.

Both FIB/SEM and TEM methods were applied to measure the thickness of the foils. The FIB/SEM measurements were carried out with FEI Helios 600 Nanolab FIB/SEM at EMSL/PNNL. Thin layers of sputtered Au were introduced on both sides of the wafer to enhance the clarity of the Si_3N_4 surfaces. Images were taken under immersion mode at a tilt angle of 52°. TEM measurements were carried out at 200 keV with a Zeiss Libra 200MC TEM/STEM within the JIAM Electron Microscopy Center at the University of Tennessee. The detector was equipped with 2 × 2 K CCD-camera. The cross-sectional TEM samples were prepared by mechanical polishing and followed by ion milling. The energy resolution was ~0.15 eV.

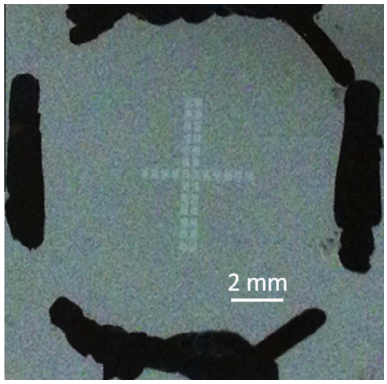

Fig. 3. A photo of a Si wafer after lateral scans by ToF-SIMS depth profiling.

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