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Sputtering yield of amorphous ^{13}C thin films under swift heavy-ion irradiation

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ABSTRACT

The sputtering yield of carbon was measured in the electronic energy loss regime by 194-MeV Au ions irradiation. Due to the overall surface contamination with hydrocarbons, the determination of sputtering yields of carbon materials is quite challenging. To separate carbon omnipresent on most surfaces from sputtered carbon, we used a carbon target consisting of 86% of ^{13}C isotope. By measuring the thickness decrease of the thin ^{13}C film by means of high resolution online Elastic Recoil Detection Analysis (ERDA), a sputtering yield of 49 ± 15 of ^{13}C per Au ion was found. Simultaneously with the sputter process, the ^{12}C content of the target increased probably due to beam-induced bond breaking of hydrocarbons attached to the surface. Our experimental results are analyzed by calculations based on the two-temperature thermal spike model.

1. Introduction

Amorphous carbon films, hydrogenated or hydrogen free, typically contain a mixture of sp^2 and sp^3 bonded carbon atoms providing excellent mechanical and tribological properties such as high hardness, low friction, and high wear resistance [1]. Ion irradiation of amorphous carbon films have been found to produce numerous effects [2] such as crystallization of the top surface to graphene [3], growth of carbon nanotubes [4], crystallographic transformations [5], changes in conductivity [6,7], and sputtering [8,9]. Sputtering is the phenomenon of particle ejection from a solid surface due to bombardment with energetic ions. Sputtering can be classified according to the mode of energy loss of the incident ion. Nuclear sputtering, dominant at ion energies of few keV/nucleon (keV/u), is a consequence of direct energy transfer from elastic collisions between the incident ion and the nuclei of the target atoms. Electronic sputtering results from the conversion of energy deposited as electronic ionization and excitation of the target atoms into atomic motion. Electronic sputtering has been reported for insulators as well as metals [10–14] with sputtering yield (atoms ejected per incident ion) depending on the sensitivity of the material to electronic energy loss. Electronic sputtering studies of carbon films have shown that the sputtering yield is quite sensitive to the thickness of the film [15], the substrate on which the thin film is deposited [16], the velocity and charge state of the ions [16], the hydrogen content [8,9,17] and the allotropic form of carbon [18].

The determination of sputtering yields of carbon films is difficult due to the general hydrocarbon contamination of surfaces. The sputter target as well as the catcher, on which the sputtered atoms are collected, may be contaminated [19,20]. In order to avoid this complication, we (i) used a thin ^{13}C film as target and (ii) monitored the thickness decrease of the target instead of applying the catcher technique [14,19–21]. Quantitative information of the thickness decrease is obtained by online high-resolution elastic recoil detection analysis (ERDA) [22,23]. Similar to previous sputtering measurements [24], the irradiation is performed with a ^{197}Au beam of 0.98 MeV/u. The sputtering yield data are analyzed and discussed within the inelastic thermal spike approach [13,14,25,26].

2. Experimental

The thin film of ^{13}C was deposited by means of electron-beam evaporation at LMU (München) at an initial vacuum of about 5×10^{-7} mbar. As substrate, we used silicon with native silicon oxide. The thickness of the ^{13}C film was measured by tapping-mode atomic force microscopy (AFM) with an ULTRAObjective

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instrument (S.I.S., Germany) at the Helmholtz-Zentrum Dresden-Rossendorf. Since the sputtering yield of carbon was found to increase with increasing hydrogen content in the film [18], it was important to quantify the hydrogen content of our film. A hydrogen depth profile of the pristine sample was obtained from $H(^{15}N,\alpha\gamma)^{12}C$ nuclear reaction analysis (NRA) with a narrow resonance at 6.385 MeV [27]. The yield of the 4.43 MeV γ-rays from the reaction for varying energy of incident $^{15}N^{2+}$ ions was measured with a bismuth germanate detector. To generate the depth scale, an average energy loss of the N ions in the carbon target of 1.82 keV/nm was used [28].

The sputtering experiments including online ERDA were performed at the tandem accelerators of LMU and TU München (Garching) [29] with 194-MeV Au ions. The vacuum in the target chamber was of the order of 10^{-7} mbar. The corresponding electronic energy loss (S_e) of Au ions when entering the carbon target is 15.3 keV/nm, as calculated with the SRIM-2010 code [28]. The beam incidence was $\alpha = 19°$ with respect to the surface of the sample. The beam current was one electrical nanoampere (nAe) of Au ions having charge state of 30+ which corresponds to 2.1×10^8 - Au ions/s. The area of the beam spot was 9.0 ± 1.5 mm^2 yielding an ion flux of about 2.3×10^{11} Au ions/cm^2 s. The recoiling atoms were monitored at a scattering angle of 37.5° using a $\Delta E - E$ gas telescope detector which has a typical energy resolution of 1% after the application of kinematic corrections [30]. The signals from the segmented electrodes of the segments of the detector were recorded event by event for higher flexibility in data handling. The two-dimensional spectra gave well separated bands of the different elements and carbon isotopes in the sample. Analysis of the ERDA spectra provided areal densities of ^{13}C as well as ^{12}C present in the irradiated film. The decrease of the areal density as a function of ion fluence provides the sputtering yield.

3. Results

3.1. Characterization of pristine film

Fig. 1 shows a line profile of the pristine carbon film across a substrate area partially masked during film deposition. From the step height, we can deduce a film thickness of 75 ± 10 nm. The depth profile obtained by NRA is presented in Fig. 2 revealing that the hydrogen content is more at the surface as well as at a depth of 75 nm due to contaminations at the surface and at the carbon–silicon interface. Within the film, the hydrogen content is less than 1 at.%. This very small hydrogen content in our films ensures us that the measured sputtering yield originates from a carbon film. A recoil spectrum of carbon obtained from online ERDA after applying a fluence of 5.6×10^{11} ions/cm^2 is shown in Fig. 3. Quantitative analysis of the spectrum with the SIMNRA code [31] revealed that the film contained a ^{12}C isotope contribution of about 14%.

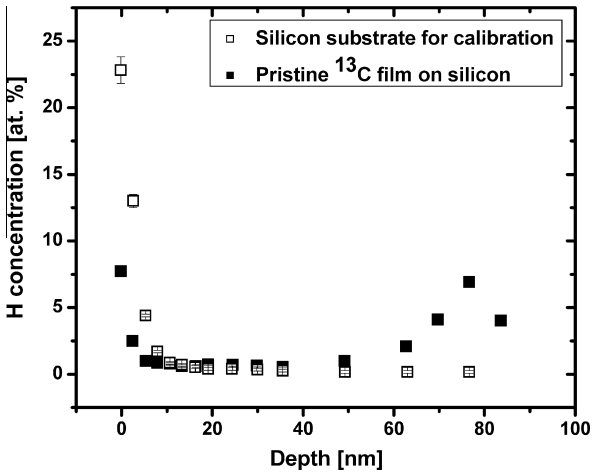

Fig. 2. Depth profiles of hydrogen in the carbon film obtained from NRA using a beam of 6.385 MeV ^{15}N ions.

3.2. Sputtering results

The recoil spectra of carbon were generated from ERDA data recorded online for different applied fluences. For quantitative analysis, the peaks of ^{12}C and ^{13}C (cf. Fig. 3) were deconvoluted, and the areal densities (in atoms/cm^2) of both isotopes were calculated from their respective peaks using the standard ERDA analysis [22]. In Fig. 4, the areal density of both isotopes is plotted as a function of the number of incident Au ions. Linear fits and extrapolation to zero beam yield an areal density of the pristine sample of $(8.5 \pm 0.1) \times 10^{16}$ ^{12}C/cm^2 and $(5.2 \pm 0.1) \times 10^{17}$ ^{13}C/cm^2. Considering the total areal density of carbon and the film thickness measured with AFM, the mass density of the film is 1.7 ± 0.2 g/cm^3.

Fig. 4(a) shows that the ^{12}C content becomes larger with increasing fluence. The rate of ^{12}C increase is equal to 51 ± 24 ^{12}C

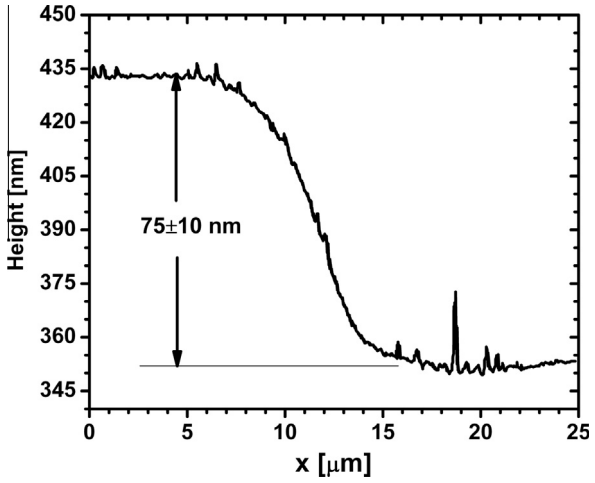

Fig. 1. Line profile of pristine ^{13}C film across a substrate area masked during film deposition. The line profile is derived from an AFM image of the sample.

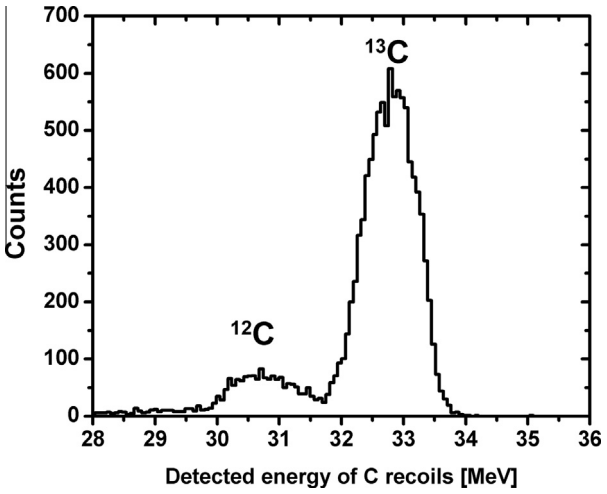

Fig. 3. ERDA recoil spectrum of carbon film showing the contribution of ^{12}C and ^{13}C. ERDA was performed with 194-MeV Au ions.