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Angular dependence of electronic sputtering from HOPG

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Abstract

We have studied the angular distribution of 120 MeV Au ion beam induced sputtering yield for three cases: from crystalline highly oriented pyrolytic graphite (HOPG) for (A) normal and (B) 70° incidence and from (C) amorphous carbon sample for normal incidence. An anisotropic distribution of sputtering is observed for HOPG samples studied with a distribution $Y = A\cos^n\theta + B\exp[-(\theta-\mu)^2\sigma^2]$. Though the over-cosine function dependence is observed for all the cases, the anomalous peak observed at 53° for normal incidence for HOPG sample is found to shift to 73° when the sample is tilted by 20°. No peak is observed in the amorphous carbon sample which further confirms that the anisotropy observed is due to the crystal structure and formation of a pressure pulse. The high exponent of over-cosine distribution of sputtering yield ($n = 3.2$–3.8) signifies formation of intense pressure pulse induced jet like sputtering.

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1. Introduction

Even though ion beams are used in many scientific and technological applications, the mechanism of transfer of energy deposited in electronic excitation to the kinetic energy of lattice atoms is still not clearly understood and is a topic of intense discussion. Historically, Fleicher et al. had proposed the 'Coulomb Explosion' model which stated that the track of damaged zone in insulators is formed due to repulsion between atoms transiently ionized by incoming ion in the solids. The Coulomb explosion model describes the coupling of electronic excitation energy into atomic motion, while according to the other competing Thermal spike model [1–5], the energy deposited by energetic ion is described by a radial temperature profile that evolves by thermal diffusion (which describes the process by transport of energy out of the heated region). Many experimental results have been explained on the basis of this model [5–9]. However, recent molecular dynamics (MD) simulations by Bringa et al. have shown that at high energy densities, models that include a thermal diffusion fail and the process instead is described by a combination of both molten track and pressure pulse [10]. The model has also been modified [11] to include mass transport induced cooling of spike leading to smaller sputtering yield. The model has recently been used by Bringa et al. [12] to effectively describe the electronic sputtering from the surface. Jakas et al. [11] have shown that the high pressure built up within the thermal spike plays an important role in sputtering process. It has been emphasized by Tombrello et al. [13] that the careful study of sputtering from surface is expected to throw light on the mechanism as the same process should be responsible for track formation in bulk and sputtering from surface. Johnson et al. [14] explained

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the sputtering by sum of impulse model and used it to describe the angular dependence of sputtering yield as well as dependence of total sputtering yield on S_e. The model also predicts the direction of momentum impulse (and hence the exit angle), which is given by $\theta_p = \pi/4 + \theta/2$ from surface normal, where θ is the incident angle. Toulemonde et al. [15] have studied the angular dependence of sputtering yield sputtering from two insulators LiF and SiO_2. Sputtering from LiF shows a jet like component symmetric to surface normal, which is explained on the basis of gas phase induced radial pressure in the track core.

It should be noted that the sputtering from various carbon allotropes has earlier been studied [16–20], though the measurement was restricted to the total yield, assuming the yield to be isotropic in all the directions. However, we have earlier [21] studied the angular distribution of the sputtering yield from HOPG for normal incidence and showed an anisotropic distribution of sputtering yield with maximum yield centred around 52°. This angular dependence was attributed to crystal structure. In the present work, we have extended the measurements to study the angular distribution of sputtering from HOPG for non-normal incidence and also from amorphous carbon for normal incidence to further confirm the effect of crystalline structure on sputtering yield.

2. Experimental

2.1. Irradiation

The HOPG samples were irradiated with a 120 MeV Au^{+9} beam from the 15 MV Pelletron [22] at Inter University Accelerator Centre (IUAC), New Delhi. The beam was incident perpendicular to the sample and the current was measured from the sample frame. The secondary electron suppressor was not used to avoid the voltage interfering with ion paths and thus affecting the measured angular distribution. Sputtered carbon atoms were collected on 10 mm × 5 mm Si catcher foils which were kept at a distance of 5 cm from the samples. The catcher foils were mounted at angles of 10–90° from the beam direction in such a way that all the catchers were equidistant in the horizontal plane. The arrangement for mounting the catcher is described elsewhere [21]. The incident current was 1.3 particle nano-amperes (1 particle nA or pnA $= 6.2 \times 10^9$ ions/s) and the sample was irradiated with 2.9×10^{13} ions to collect sufficient carbon on catchers for ERD analysis. Since the exposure time was long, the beam spot was changed after attaining overlapping fluence on the sample. Each catcher subtends an angle of 6°, with an error of ±3° in the angular measurement, and an equal contribution is expected from the error due to beam spot size (2 mm × 5 mm).

After the irradiation was complete, a 20 nm Al layer was deposited on the catchers to avoid the collected carbon getting sputtered during the subsequent ERDA study. This was important as the same 120 MeV Au beam was used for sputtering carbon from HOPG as well as for ERDA study. A dummy catcher was also placed in the chamber so that we can measure only the sputtered carbon, as contributions from hydrocarbons and other contaminants present in the chamber are also expected. The dummy catcher was placed in the chamber on the outer wall in such a way that it does not see the sputtered atoms. This catcher went through all the processes, including the Al layer deposition and the carbon collected on this dummy was measured to find the carbon contribution from various contaminants on the catchers. This contribution was subtracted while calculating the actual quantity of carbon collected on each catcher during sputtering measurement. To observe the effect of crystal structure on distribution of sputtering yield, an HOPG sample was tilted by an angle of 20° and experiment was repeated. The experimental conditions were same as above except that the sample was kept at an angle of 70°. The sample was irradiated for a fluence of 2.9×10^{13} ions/cm^2. To observe the effect of crystal structure on distribution of sputtering yield an amorphous carbon sample, prepared by thermal evaporation was used. The experimental conditions were same as for HOPG sample with normal incidence. The sample was irradiated with 3.2×10^{13} ions.

2.2. ERDA study

The carbon collected on the catcher foils is analyzed using the ERDA technique using 120 MeV Au beam at IUAC. The sample was kept at an angle of 20° with respect to the beam direction and the recoils were detected at an angle of 45°. The recoils were detected using a large area gaseous $dE - E$ detector. Isobutane gas at 40 torr pressure was used in the detector. The ERD spectra for ten catchers was recorded and the absolute quantity of sputtered carbon on each catcher was obtained from the total counts in the carbon band of the two dimensional spectra.

The sputtering measurements are sensitive to charge state of the incident ion. For 120 MeV Au ions, the equilibrium charge state is $\sim 28^+$. However for optimum beam current, in the present experiment, we have used Au^{9+} beam and hence the observed yield may be 50% of yield for equilibrium charge state [23]. We would like to point out that we have compared the sputtering yield for the three cases with same charge state and hence the charge state of Au ions will not have any bearing on the conclusions reached in this study.

3. Results and discussion

The areal density of carbon collected for HOPG sample with normal incidence on the catchers is shown in Fig. 1a. Since the sputtering yield is angle dependent, the value of total carbon collected by all the catcher foils is calculated by integrating the measured yield at a given angle. Since our measurements are restricted to only one azimuthal plane we have assumed the distribution to be isotropic in