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Sputtering of oxygen ice by low energy ions

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article info abstract

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Naturally occurring ices lie on both interstellar dust grains and on celestial objects, such as those in the outer Solar system. These ices are continuously subjected to irradiation by ions from the solar wind and/or cosmic rays, which modify their surfaces. As a result, new molecular species may form which can be sputtered off into space or planetary atmospheres. We determined the experimental values of sputtering yields for irradiation of oxygen ice at 10 K by singly (He⁺, C⁺, N⁺, O⁺ and Ar⁺) and doubly (C²⁺, N²⁺ and O²⁺) charged ions with 4 keV kinetic energy. In these laboratory experiments, oxygen ice was deposited and irradiated by ions in an ultra high vacuum chamber at low temperature to simulate the environment of space. The number of molecules removed by sputtering was observed by measurement of the ice thickness using laser interferometry. Preliminary mass spectra were taken of sputtered species and of molecules formed in the ice by temperature programmed desorption (TPD). We find that the experimental sputtering yields increase approximately linearly with the projectile ion mass (or momentum squared) for all ions studied. No difference was found between the sputtering yields for singly and doubly charged ions of the same atom within the experimental uncertainty, as expected for a process dominated by momentum transfer. The experimental sputter yields are in good agreement with values calculated using a theoretical model except in the case of oxygen ions. Preliminary studies have shown molecular oxygen as the dominant species sputtered and TPD measurements indicate ozone formation. © 2015 Elsevier B.V. All rights reserved.

1. Introduction

Studies of ion-induced processing of astrophysical ice analogues in the laboratory are relevant to a variety of different environments, such as the icy mantles of dust grains in the interstellar medium and protoplanetary discs, together with the surfaces of Solar system objects such as comets, Centaurs, Kuiper belt objects, and the icy satellites of the outer planets. Sputtering by ion impact can give rise to changes in the chemical, physical and optical properties of the ice as a result of both elastic and inelastic collisions. In the sputtering process, material is ejected from the surface which may assist in the development of thin atmospheres such as those found around Pluto and the icy moons of Jupiter and Saturn. An extreme example is Europa, where O_2 on the trailing hemisphere of Jupiter's moon Europa is sputtered by lowenergy ions to become the dominant component of the moon's atmosphere and neutral gas torus [1–[3\]](#page--1-0), together with causing the hemispherical colour variations.

Sputtering also plays a role in the chemical alteration of distant comet surfaces, and contributes to the formation of the first coma as these objects approach the sun. Recently, the signature of this process was recorded by the ROSINA mass spectrometers on-board the Rosetta spacecraft at comet 67P/Churyumov–Gerasimenko. Small amounts of

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refractory elements such as Na and Si are believed to have been generated by Solar wind sputtering of dust grains on the surface of the nucleus [\[4\].](#page--1-0)

 $O₂$ ice is not a major component of the observed astrophysical ices inventory but $O₂$ ice processing by the slow component of the Solar wind and cosmic rays plays an important role in the formation of other species such as ozone [\[5\],](#page--1-0) water and various carbon-oxides [\[6\].](#page--1-0) Whilst there have been many sputtering studies using water ice as a target (see [\[7\]](#page--1-0) and references within), sputtering of oxygen ices has seen fewer investigations. Given that cosmic ray impacts onto molecular oxygen on dust grain surfaces is one possible pathway in the formation of water (e.g., [\[6\]\)](#page--1-0), the associated loss pathway of sputtering warrants further investigation. Depending on the energy of the incoming projectile ion, experimental sputtering can be divided into two main categories: sputtering by low energy ions (a few keV) and sputtering by high energy ions ($>$ 30 keV, up to \sim 3 MeV). At energies of a few keV ions primary lose energy in 'nuclear' ballistic collisions, where target molecules are excited vibrationally, rotationally and translationally. Above 30 keV energy is lost primarily in electronic excitation of target molecules by, e.g., ionisation. In between these two energy ranges neither of these two energy loss mechanisms dominates.

Sputtering of oxygen ice by low energy (4-10 keV) H^+ , H $_2^+$, and H $_3^+$ and high energy ions He⁺ and H⁺ (up to 3.5 MeV) has been intensively studied by Ellegaard et al. [\[8,9\]](#page--1-0), and references within. They concluded that oxygen ice sputters more efficiently than nitrogen ice by a factor

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of almost 2, and that the sputter yields are proportional to the electronic stopping power, i.e., the energy lost by the projectile ion due to inelastic collisions with bound electrons as it penetrates the ice. The effect of multiply-charged ions on ion-induced sputtering of $O₂$ has been neglected so far in the literature, with the only study at low energy for oxygen ices by Gibbs et al. [\[10\]](#page--1-0) on oxygen ice irradiated at MeV energies by He^{2+} and He^{+} ions. They concluded that the yields are a factor of 1.2 higher for He^{2+} ions.

The present work is a laboratory study of sputtering of oxygen ice by 4 keV singly and doubly charged C, N, and O ions and singly charged He and Ar. There have been only a few laboratory experiments on sputtering by low energy non-noble gas ionic species such as C, N and O, which have the potential to be incorporated in new molecules in surfaces and even fewer experiments for sputtering by multiply charged ions.

2. Experimental details

The experimental arrangement used in the present work is shown schematically in Fig. 1. An Ultra High Vacuum (UHV) chamber was coupled to a low energy ion accelerator [\[11\]](#page--1-0) equipped with a 10 GHz Electron Cyclotron Resonance (ECR) ion source [\[12\].](#page--1-0) Singly and doubly charged ions of ¹³C, ¹⁴N, ¹⁶O and singly charged ions of ⁴He and ⁴⁰Ar, at 4 keV, were focused and directed into the experimental chamber (which operated at a base pressure $\sim 1 \times 10^{-9}$ mbar). Ion beams were collimated by three 4 mm diameter apertures separated by 50, 38 and 26 mm respectively from the substrate providing an ion beam of 4 mm in diameter with less than 1% increase in diameter for the maximum current density used at the ice surface. A potassium bromide (KBr) substrate, 20 mm in diameter and 2 mm thick, was clamped to an earthed, temperature controlled sample holder (A.S. Scientific) surrounded by a gold plated copper radiation shield.

The substrate temperature was controlled to within 0.1 K at temperatures from 8 to 300 K using a calibrated silicon diode. A slot in the radiation shield 15 mm high and with a total angular width of 120° enables ion beam access to the deposited ices; the slot gives access of $\pm 60^{\circ}$ with respect to the normal value of the substrate surface in the radiation shield. The radiation shield temperature was measured throughout the experiment by a diode, and was found to be constant at a value of 48 K during the irradiation and heating procedure. The

Fig. 1. Schematic diagram of the experimental arrangement with 1) ion irradiation part, 2) KBr substrate, 3) radiation shield, 4) vapour deposition nozzle, 5) and 6) thickness monitor comprising (5) a 405 nm diode laser and (6) a photodiode detector and 7) line of sight quadrupole mass spectrometer. Fig. 2. Reflection and refraction model for an oxygen ice film deposition on a KBr window.

cold-head was rotatable through 360° via a differentially pumped rotary stage. The substrate could be rotated to any desired position during the experiment. Additionally, as shown schematically in Fig. 1 a quadrupole mass spectrometer (QMS; model HAL-201 from Hiden Analytical) could be used to monitor species emanating from the sample either from sputtering or thermal desorption.

The ice films were deposited at a pressure of 1.6×10^{-7} mbar with the thickness measured by laser interferometry using a 405 nm diode laser and a photodiode detector. During deposition, the cold-head assembly was rotated such that the vapour deposition nozzle was directed normal to the substrate surface, at a distance of 25 mm, with the laser and detector at 45° with respect to the substrate normal. Oxygen ice films of up to 258 nm were deposited onto the substrate which was held at 10 K.

Fig. 2 shows a schematic diagram of the reflection and refraction for the oxygen ice film deposited on a KBr window. θ_0 is 45° and represents the angle of incidence of the laser light with respect to the surface normal, and θ_1 is the angle of refraction at the vacuum ice interface calculated from Snells Law (1). n_0 is the vacuum refractive index and n_1 the oxygen ice refractive index.

$$
n_0 \sin \theta_0 = n_1 \sin \theta_1 \tag{1}
$$

The intensities I_1 to I_5 can be calculated from the reflection and refraction laws. The model fit equation is expressed as

$$
PS = C \left(I_1 + T^{2d/\cos \theta_1} \times I_5 \cos \frac{\pi d}{\cos \theta_1} \right) \tag{2}
$$

where PS is the photodiode signal in volts, I_1 is the reflection intensity from vacuum to the ice and I_1 the refracted intensity after the light passes through the ice and is reflected back. In the model a value 1.285 for the refractive index of the ice gives the best fit. C is a constant related to the photodiode efficiency and T is the transmission coefficient of the ice at 405 nm. In Eq. (3) , m is an integer, where odd values of m correspond to maxima (constructive interference) and even values of m correspond to minima (destructive interference) at intervals of 86 nm for this wavelength.

$$
d = m\frac{\lambda}{4}\cos\theta_1\tag{3}
$$

The ice density was calculated using the Lorentz–Lorenz relation [\[13\]](#page--1-0)

$$
L \times \rho = \frac{n^2 - 1}{n^2 + 2} \tag{4}
$$

where L is the Lorentz–Lorenz factor, n is the refractive index of the ice and ρ the density of the ice. A value of $L = 0.1294 \text{ cm}^3 \text{ g}^{-1}$ has been used in the present work $[13]$ and we assume that L is constant in the

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