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First experimental data of sulphur ions sputtering water ice

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ABSTRACT

This paper presents the first experimental sputtering yields for sulphur ions with energies between 10 keV and 140 keV irradiating water ice films on a microbalance. The measured sputtering yields exceed theoretical predictions based on other ion species by a factor of 2 to 3 for most energies. Moreover, the sputtering yield of SF^+ molecules is compared to the yield of atomic species S^+ and F^+. As found for atomic versus molecular oxygen, the sputtering yield caused by molecules is two times higher than expected. Finally, the implications of the enhanced sulphur sputtering yield for Europa's atmosphere are discussed.

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1. Introduction

When water ice is irradiated with energetic ions, the energy transferred from the impactor to the ice may eject particles from the surface. This process, termed sputtering, has been studied for several ion species and energy ranges under laboratory conditions over the past decades (e.g., Haring et al., 1984b and accompanying papers, (Baragiola et al., 2003; Farenzena et al., 2005; Famá et al., 2008; Johnson et al., 2009; Cassidy et al., 2013; Muntean et al., 2016; Galli et al., 2017). Such results are needed to check theoretical models of sputtering (Sigmund, 1969; Johnson, 1989; Cassidy and Johnson, 2005) and relate them to observations (Hall et al., 1995; Hansen et al., 2005; Roth et al., 2016) and models (Shematovich et al., 2005; Smyth and Marconi, 2006; Plainaki et al., 2012; Vorburger and Wurz, 2018) of sputter-induced atmospheres of icy moons such as Europa.

Sputtering may proceed in a straight-forward manner, i.e., the ion directly knocking out one or several water molecules, or it may be a two-stage process with the irradiation first causing chemical reactions inside the ice (so-called radiolysis (Haring et al., 1984a; Johnson et al., 2004; Cassidy et al., 2010)) and subsequently releasing the radiolysis products from the surface. The sputtering yield denotes in both cases the number of water molecules or equivalents (if H_2O reacted to H_2 and O_2, for instance) leaving the ice per impacting particles. Knowing this yield and the chemical and energetic composition of the ejecta over a wide range of parameters is important to understand any ice-covered celestial body. The astrophysical application we are most interested here is Europa, one of the icy moons of Jupiter. For these bodies, the sputtering yields and the plasma environment determine the density and composition of their atmospheres (see Johnson et al. (2004) for a review).

For this study, we sputtered thin water ice films with sulphur ions and sulphur-bearing molecules from a microbalance. This is the most common experimental method used so far (see for example Baragiola et al., 2003; Teolis et al., 2005; Famá et al., 2008; Muntean et al., 2016; Galli et al., 2017). However, to our knowledge, sulphur ions were never used before in such experiments as sulphur is chemically reactive and can corrode surfaces in vacuum chambers. Argon was the species closest in mass to sulphur so far, for which experimental results exist (see compilations by Johnson et al., 2009; Cassidy et al., 2013). Knowing the sputtering yield of S^+ on water ice is highly relevant for Europa's atmosphere: S^+ ions are one of the three most frequent ion species in the plasma environment around Europa (Paranicas et al., 2002) because of its volcanically active neighbour Io.

After a recapitulation of the theory of ice sputtering (Section 2), we describe the experiment set-up in Section 3. We then present the sputtering results for S^+, F^+, and SF^+ molecules (Section 4). The paper is completed with a discussion on the implications of these results for Europa and for future laboratory work (Section 5), followed by the conclusions (Section 6).

2. Theory

We will compare our new experimental results for S^+ and F^+ to two widely used semi-empirical formulae for ion sputtering yield derived by Famá et al. (2008) and Johnson et al. (2009). These formulae are based on previous experiments with other ion

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species irradiating dense water ice films. For ion energies below 10 keV, the sputtering yield can be described by a cascade of elastic collisions, whereas the so-called electronic sputtering dominates at higher energies. Eq. (1) by Famá et al. (2008) is more accurate for lower energies; for energies above 100 keV, Eq. (2) by Johnson et al. (2009) offers a better fit to experimental data for H^+, He^+, N^+, O^+, Ne^+, and Ar^+ ion beams (Cassidy et al., 2013):

$$Y(E, m_1, Z_1, \theta, T) = \frac{1}{U_i}\left(\frac{3}{4\pi^2 C_0}\alpha S_n + \eta S_e^2\right)\left(1 + q_i \exp\left(-\frac{E_a}{k_B T}\right)\right)\cos^{-f}(\theta) \qquad (1)$$

Eq. (1) quantifies the sputtering yield as a sum of nuclear and electronic sputtering, described by the nuclear stopping cross section $S_n(E, m_1, Z_1) = dE_n/(Ndx)$ and the electronic stopping cross section $S_e(E, m_1, Z_1)$. The yield in Eq. (1) depends on energy E, mass of impactor m_1, atomic number of impactor Z_1, the incidence angle θ relative to the surface normal, and surface temperature T. The temperature-dependent term with the activation energy E_a (Reimann et al., 1984) becomes dominant above $T = 120$ K and is due to radiolysis and subsequent release of H_2 and O_2 (Johnson et al., 2004; Famá et al., 2008; Teolis et al., 2009). This contribution makes up only 10% of Y at 90 K (Eq. (1)); it rises to 20% at 100 K once the ice has been saturated with $\sim 10^{15}$ ions cm^{-2} (Teolis et al., 2005). For U_i, the sublimation energy of water (0.45 eV) is assumed. The effective cross-section for low energy recoils, $C_0 = 1.3$ Å^2, the activation energy, $E_a = 0.06 \pm 0.01$ eV, and $q_i = 220$ are constants (Famá et al., 2008). The parameter describing the angular dependence calculates to $f = 1.75$ for S^+.

By comparison, Johnson et al. (2009) propose the following empirical formula for the sputtering yield (the angular and temperature dependence are identical to Eq. (1)):

$$Y(E, m_1, Z_1, \theta, T) = 1/(1/Y_{low} + 1/Y_{high})\left(1 + q_i \exp\left(-\frac{E_a}{k_B T}\right)\right)\cos^{-f}(\theta) \qquad (2)$$

whereby Y_{low} and Y_{low} stand for:

$$Y_i = Z_1^{2.8} C_1 \left(\frac{v}{2.19 \times 10^6} Z_1^{-1/3}\right)^{C_2}, \text{ with ion velocity } v = \sqrt{2E/m_1} \qquad (3)$$

The fit parameters are $C_1 = 4.2$ and $C_2 = 2.16$ for Y_{low} and $C_1 = 11.22$ and $C_2 = -2.24$ for Y_{high}.

3. Experiment set-up

The same microbalance set-up was used as in our previous ice film experiments (Galli et al., 2017): We background deposited deionized water vapour via a needle valve and a capillary onto the cooled surface of a microbalance in a vacuum chamber. The sensitivity of the microbalance was 1.61×10^9 Hz g^{-1} according to calibration in 2014 performed by the manufacturer (gold-coated 15 MHz quartz crystal, manufacturer: QCM Research). The surface of the microbalance was 45° or 60° tilted with respect to the incoming ion beam. Under these conditions and temperatures around 90 K, most of the deposited ice will remain amorphous throughout the experiments and the porosity will vary between few % (Famá et al., 2008) and 25% (Mitchell et al., 2017). If the bulk density is 0.9 g cm^{-3}, one monolayer of H_2O on the microbalance corresponds to a frequency shift of 14 Hz. The H_2O partial pressure in the chamber ranged from 3×10^{-8} to 4×10^{-7} mbar during vapour deposition; the ice film accretion rate increased linearly with that partial pressure from 0.2 to 5 Hz s^{-1}. Within this range, the deposition rate did not notably affect the measured sputtering yields. For irradiation experiments, the ice film thickness ranged from 40 to 200 nm and the residual water pressure in the vacuum chamber was only on the order of 10^{-9} mbar.

To create an ion beam, we ionized SF_6 gas and accelerated the ion species with an electron-cyclotron-resonance ion source (Marti et al., 2001). The ion source produced many different species from the parent molecule SF_6, but only S^+, F^+, S^{2+} and SF^+ turned out to have a sufficiently high beam current to create a detectable sputtering signal when the ion beam was directed at the water ice film on the microbalance. The beam currents reached 0.1 to 1.0 nA, which corresponded, at a beam diameter of 0.3 cm, to $(0.9 \ldots 9) \times 10^{10}$ ions cm^{-2} s^{-1}. To interpret the results for the SF_6 fragments and to verify the microbalance sensitivity, we also irradiated the microbalance with O^+ ions whose sputtering yield is well known from previous studies (Shi et al., 1995; Baragiola et al., 2003).

4. Results

4.1. Accuracy of results

Before we discuss the results for the hitherto unknown sputtering yields of S^+, F^+, and SF^+, let us first assess the general accuracy of our experiments using oxygen as a reference. Oxygen sputtering yields have been measured numerous times by us and other research groups whereby Eq. (1) fits most previous laboratory experiments within 30% relative uncertainty (Famá et al., 2008). We therefore collected all O^+ yield results measured at an impact angle of 45° and 10, 30, and 50 keV energy over the last 1.5 years in our facility. This data set was accumulated during five different measurement series separated by several weeks or months. The first part of this data set covering the year 2016 was presented in Galli et al. (2017); here we added the measurements from 2017. We normalized all data (obtained at temperatures between 89 and 101 K) to the same temperature $T = 90$ K assuming the temperature dependence in Eqs. (1) and (2). Apart from temperatures also vacuum pressure, vapour deposition rate, ice film thickness, and irradiation duration varied. Moreover, we used two different microbalances of the same type (see Section 3).

The average sputtering yields derived from this comprehensive data set compared to the Y_{th} predicted from Eq. (1) (with $T = 90$ K) as follows: $Y = 44 \pm 13$ (14 data points) vs. $Y_{th} = 27$ at 10 keV, $Y = 73^{+14}_{-25}$ (7 data points) vs. $Y_{th} = 62$ at 30 keV, and $Y = 111^{+15}_{-33}$ (5 data points) vs. $Y_{th} = 105$ at 50 keV. The experimental error bars denote the ranges between average and most extreme positive and negative outlier.

These results are important in two respects: First, the experimental values are reproducible within 30% or better on the long run. During one measurement series, the scatter usually was on the order of 10% (Galli et al., 2017). Second, the oxygen results agree with Eq. (1), which reproduces previous results from other groups within 30% (Famá et al., 2008). We will therefore attribute an uncertainty of 30% to our experimental yields for other ion species in the following section.

4.2. Results for S^+, F^+, and SF^+

105 individual irradiations with S^+, SF^+, S^{2+}, and F^+ ions hitting the ice film were performed during two different measurement series of six days in total. 11 of the S^+ irradiations took place the same day when also O^+ sputtering yields were measured for cross-calibration (see Section 4.1). The median temperature of the water ice film during the measurements was 90 K with extremes of 89 K and 101 K. For evaluation, individual sputtering yields obtained at a temperature other than 90 K were normalized to $T = 90$ K based on Eqs. (1) and (2). At the given temperature range this corresponds to a modification by a factor of 1.1 at most. An individual irradiation lasted between 1 and 30 min; Fig. 1 shows as an example the frequency response of the microbalance (in Hz

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