

## Classical dynamics study of the ion bombardment of ice

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A classical dynamics model is used to simulate the bombardment of H<sub>2</sub>O ice by O<sup>+</sup> ions at energies between 23 and 115 eV. These energies are appropriate to describe the erosion processes occurring on the inner moons of Saturn. The total erosion yields vary between 0.83 and 2.28 H<sub>2</sub>O equivalents per incident ion in the energy range investigated. The majority of the ejected material is intact H<sub>2</sub>O molecules which have very little internal energy. Clusters of the type (H<sub>2</sub>O)<sub>n</sub> are observed in the calculations. Clusters of the form (H<sub>2</sub>O)<sub>n</sub>H<sup>+</sup>, however, are not observed. This is consistent with experimental results of others on cluster and fragment formation. Predictions made by a collision-cascade model for the absolute erosion rates of ejected species are shown to be inappropriate for these low impact energies.

## I. INTRODUCTION

Many of the moons of the outer planets in our solar system are continually subjected to bombardment by energetic heavy particles. For example, the icy moons of Saturn are bombarded by O<sup>+</sup> ions trapped in the planet's magnetosphere.<sup>1</sup> This natural event is similar to that occurring in a number of earth-bound laboratory experimental techniques such as secondary-ion-mass spectrometry (SIMS), fast atom bombardment mass spectrometry (FABMS), and a vast array of sputtering experiments. Unlike the conditions of a laboratory experiment, however, it is possible that the particles ejected from a moon undergo a number of subsequent interactions. If the particles lack the necessary kinetic energy to escape the moon's gravitational attraction, they will enter into a ballistic trajectory and be redistributed elsewhere on the planetary surface. This sputter-transport mechanism has been used to interpret the latitudinal variation in the reflectance spectra of the bombarded moons.<sup>2</sup> Alternately, species ejected with sufficient energy can escape the gravitational field of the moon. These particles will then be ionized by solar photons or magnetosphere electrons and become part of the collection of ions trapped in the planet's magnetosphere.<sup>3</sup> Clearly, the details of such a satellite wide sputtering mechanism depend very strongly on the energy distribution, angular distribution, and absolute yield of the sputtered products.

The general interactions responsible for the sputtering of ice can be divided into two regimes. The high-energy primary particles fall into the first regime where the interactions are due to electronic processes. This regime has been studied both experimentally and theoretically by Lanzerotti *et al.*,<sup>4</sup> where it has been shown that for primary particles with energies of 10 keV to several MeV, the sputtering of water molecules is proportional to the square of the electronic stopping power. For lower energy primary particles, the momentum exchange is due to elastic nuclear collisions. Bar-Nun *et al.* have experimentally studied the transition between the two regimes and indicate that for Ne<sup>+</sup> ions incident on ice with energies in the

0.5–6 keV region, the interactions are exclusively due to nuclear collisions.<sup>5</sup>

In order to obtain a detailed understanding of the sputtering of molecular solids in the low-energy primary-ion regime, we have developed a classical dynamics model of the processes. This procedure has been used successfully to predict observables from SIMS experiments on clean metals and metals with atomic and molecular adsorbates.<sup>6</sup> These calculations are especially appropriate for the erosion of the Saturnian moons since an estimate of the total amount of material removed is required. Most experimental configurations do not measure the yield of all types of ejected species (e.g., ions and neutrals) simultaneously.

There are several questions that are relevant to the astrophysical problem. (i) What is the absolute erosion rate for each of the Saturnian moons given in Table I? Note that the energy of the bombarding O<sup>+</sup> ion for each moon depends on the distance of the satellite from the planet. (ii) What is the energy distribution of the ejected species? (iii) What is the angular distribution of the sputtered material? (iv) What is the nature of the ejected material (e.g., H<sub>2</sub>O, OH, H, etc.)? (v) What is the fate of the impacting ion?

In addition to the astrophysical questions, it is of interest to examine the mechanisms of ejection for a molecular solid. Previous simulations of the ion-bombardment process have been performed on clean metals or atomic and molecular adsorbates on metal surfaces. There are several differences between molecular and atomic solids which are important for sputtering processes. Most importantly, molecular solids contain internal chemical structure which can absorb some of the energy of the primary ions into internal energy of individual molecules. This internal excitation can be estimated from the final positions and momenta of the atoms comprising the molecule. Other differences involve the possibility that molecular solids maintain long-lived localized charges in the bulk and that the heats of sublimation are generally lower for molecular solids than for metals.

In this study the theoretical treatment of the ion bom-

TABLE I. Moons of Saturn. Data taken from Ref. 13.

Moon	Radius (km)	Orbital radius ( $10^3$ km)	H <sub>2</sub> O escape energy (eV)	Incident O <sup>+</sup> energy (eV)
Mimas	180	185	0.00237	23
Enceladus	250	238	0.00417	59
Tethys	530	295	0.018	115
Dione	560	377	0.023	224
Rhea	765	527	0.040	504

bardment of molecular solids and the implications for terrestrial experiments is emphasized. The analysis of the results for the astrophysical problems of erosion and sputter transport will be treated separately. The calculational procedures are described in Sec. II and the results are presented in Sec. III.

## II. DESCRIPTION OF THE CALCULATION

The erosion of ice is modeled by a classical dynamics procedure which has been previously developed for the interpretation of ion-solid collision experiments.<sup>6</sup> The solid is modeled by an array of water molecules arranged tetrahedrally in a diamond lattice. The O<sup>+</sup> ions are incident perpendicular to the surface with kinetic energies chosen to simulate the extraterrestrial conditions given in Table I. The atoms in the crystal and the bombarding ion are allowed to interact classically and their motion is followed by integrating Hamilton's equations of motion until the kinetic energy of each particle in the crystal is below a certain minimum threshold energy (i.e., until no more species can eject). The simulation is then terminated and the positions and momenta of all the particles above the surface are analyzed to determine the ejection yields, energy and angular distributions, and amount of internal energy in each sputtered molecule.

Ice exhibits a multitude of crystalline structures as well as an amorphous phase. In this work we are interested in ice formed under the conditions of low temperature and pressure, where either the *Ic* structure (where the oxygen atoms are arranged in a diamond lattice) or amorphous ice is prevalent.<sup>7(a)</sup> Since the exact structure of the ice in the Saturnian moons is not known and experimental results on the sputtering of ice have been shown to be unaffected by the relative amount of amorphous and cubic ice bombarded,<sup>5</sup> a crystallite with oxygen atoms arranged in a diamond structure is used in the calculations. The hydrogen atoms are arranged according to the Bernal-Fowler rules, where the identity of each water molecule is retained in the crystal.<sup>7(b)</sup> This is consistent with the *Ic* structure and also facilitates the formation of gas-phase single molecules and clusters composed of single molecules as seen experimentally.<sup>5</sup> Disordered systems are not treated. Each intramolecular oxygen-hydrogen bond then points toward the oxygen atom of another molecule so that each oxygen atom has two intramolecular and two intermolecular hydrogen atom neighbors, except of course, at the surface and on the edges. The intramolecular O-H and inter-

molecular O-O distances are initially set at 0.992 and 2.74 Å, respectively. The initial intramolecular H-O-H angle has the ideal tetrahedral value (109.47°). Preliminary testing on the {100} and {110} faces indicated that the total yield and center-of-mass energy distributions for this system are relatively insensitive to the crystallographic orientation. Therefore, only one exposed face, the {110} face, is considered and all results reported here are for trajectories performed with that orientation. The crystallite used is composed of four layers, where the top and the third layers each contain 50 molecules and the second and fourth layers contain 32 molecules (Fig. 1).

The choice of potential energy interactions among the particles is constrained by several conditions. As discussed above, the identity of each water molecule is retained in the crystal. Thus, the potentials are separated into intermolecular and intramolecular interactions. Since the total amount of erosion of the surface and the amount of fragmentation of the ejected species is of interest, intramolecular potentials that reasonably describe the dissociation of H<sub>2</sub>O are required. While several potentials for nonrigid water molecules in the bulk phase are available from the literature, none satisfy this fragmentation criteria. Consequently, the potential surface of Schinke and Lester<sup>8</sup> which had originally been developed to study gas-phase reactions but also gives good agreement with equilibrium parameters for isolated water molecules is used. Unfortunately, this potential includes a rather unwieldy three-body term which is necessary to insure a correct equilibrium configuration for the H<sub>2</sub>O molecule. Inclusion of this term, while we feel necessary at least initially, increases the amount of computer time needed for each trajectory, thus limiting the number of trajectories that can economically be determined. The computer time for each ion impact is ~4 h on a Floating Point System FPS-5205 Array Processor.

The choice of intermolecular potentials is, in comparison to that of intramolecular interactions, rather arbitrary. The constraint applied here is that the forces be directed between atoms in the molecule so that the motion of the atoms can be straightforwardly determined. Several adequate potentials for ice are available in the literature and detailed comparisons among them have been made.<sup>9</sup> The transferable intermolecular potentials (TIP's) developed by Jorgensen<sup>10</sup> have been chosen because of their simple form. The only modification made is to include a short-range oxygen-hydrogen repulsive term. The added term used is the same one that Pettit

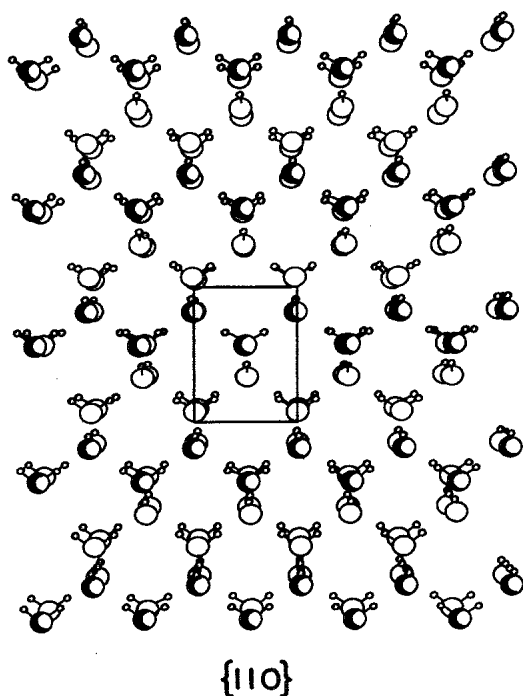


FIG. 1. The crystal face bombarded. There is slight perspective in the drawings. The first-layer oxygen atoms are shaded. The rectangular region in the center is the irreducible symmetry zone in which the ion bombards.

and Rossky have included in their simulations with the TIP's.<sup>11</sup>

The potential used for the impinging ion with the oxygen and hydrogen atoms is of the Moliere form, with the screening length given by the Firsov value.<sup>12</sup> This potential form and screening length is appropriate for describing nuclear collisions. No attempt was made to fit this to the repulsive terms in the TIP's potential, which were determined by Jorgensen on the basis of the equilibrium structure and energetics of the water dimer and of liquid water.

The composite potential has the following characteristics. The energy to remove one water molecule from the solid is 0.5 eV. The energy to completely dissociate a H<sub>2</sub>O molecule into atomic fragments is 8.95 eV. The energy to remove one H atom from the molecule is ~4.9 eV. Since the atoms that were originally in a given molecule are prescribed, formation of H<sub>2</sub>O molecules from dissociated fragments is neglected. This omission should not be too restrictive, since at these low primary-ion energies the average yield of ejected dissociated molecules is less than one per incident ion. In addition, since each ion impacts a fresh target, the damaged targets where recombination processes might occur are unimportant.

For each trajectory the incident O<sup>+</sup> ion is initially ~3 Å above the surface and is aimed within the irreducible symmetry zone of the crystal face shown in Fig. 1. Three different initial energies of the incident ion are used in order to reproduce conditions found on the inner moons of Saturn (see Table I). Each trajectory is followed until the kinetic energy of each of the particles remaining within

the crystal is below a threshold energy. This energy is 0.5 eV if the atoms are in the top two layers of the crystal. A cutoff value of 1 eV for all particles deeper in the solid does not affect the results but does decrease the required computer time. Typical time steps are on the order of 0.3 fs (1 fs = 10<sup>-15</sup> s) and the elapsed time for each trajectory is on the order of 150 fs. The final positions and momenta of particles ejected from the surface are analyzed for the total yield of sputtered particles, degree of fragmentation of the water molecules, and the energy and angular distributions of the ejected molecules.

### III. RESULTS

Given in Table II are the calculated yields of various ejected species as a function of O<sup>+</sup>-ion energy. Calculations were not performed at primary-ion energies of 224 and 504 eV because preliminary trajectories indicated that a larger crystal size would be needed and the computer time would be astronomical. In addition, it is believed that most of the sputtering occurring on Rhea is due to much more energetic (~50 keV) protons and O<sup>+</sup> ions.<sup>13</sup> Several trends are apparent from these results. First, most of the sputtered material consists of intact single H<sub>2</sub>O molecules. Also, the total yield of sputtered material increases with increasing primary-ion energy. Finally, virtually all of the incident O<sup>+</sup> primary ions implant rather than reflect from the surface, with the frequency of implantation increasing with increasing incident energy.

A direct comparison between our results and experimental results suffers from several difficulties. First, most of the experimental results available in the literature have been performed under different conditions than we have simulated here. The most important differences are that the experiments have used higher-energy primary ions and high ion dose conditions, where an already damaged crystal was rebombarded. This has been shown to eject species that had been produced (but not sputtered) during a previous bombardment event.<sup>14,15</sup> In addition, several of the experimental product yields have been time and temperature dependent.<sup>5</sup> This again is probably due to the subsequent removal of species created during a previous bombardment.

The experiment which is most relevant to this work is that of Bar-Nun *et al.*<sup>5</sup> In their experiment ice was bombarded by Ne<sup>+</sup> ions in the range of 0.5–6 keV and absolute product yields were measured for substrate temperatures between 54 and 140 K. Several interesting results were reported. First, they found that the yield of H<sub>2</sub>O molecules was constant at around 10 per incident ion in the primary-ion energy range between 2 and 6 keV and then dropped off at lower energies. This yield extrapolates reasonably well to our results. Furthermore, it was found that the yields of intact H<sub>2</sub>O molecules and H single atoms were substrate temperature independent while the number of H<sub>2</sub> and O<sub>2</sub> molecules detected was strongly temperature dependent. They speculated that the sputtered H<sub>2</sub>O molecules and H atoms were originally surface species and that the H<sub>2</sub> and O<sub>2</sub> molecules were formed further in the bulk ice and had to diffuse to the surface to be sputtered. The rate of this diffusion was presumed to

TABLE II. Calculated yields.

	No. of impact points										Total equivalents <sup>c</sup> of H <sub>2</sub> O removed per incident ion	Haff <sup>d</sup> estimations
		H <sub>2</sub> O	OH	O <sup>a</sup>	H	O <sup>+</sup>	(H <sub>2</sub> O) <sub>n</sub>					
							n=2	3	4	5		
23 eV {110}	96	69(0.72) <sup>b</sup>				16	4	1			0.83	1
59 eV {110}	96	137(1.43)			4	3	8	3		2	1.83	3
115 eV {110}	96	144(1.50)	5	1	16	1	16	8	3		2.28	5

<sup>a</sup>O refers to oxygen atoms originally in a water molecule and O<sup>+</sup> refers to the incident ion.

<sup>b</sup>The number in parenthesis is the yield of single H<sub>2</sub>O molecules ejected per incident ion.

<sup>c</sup>Equivalents are defined here as the total mass ejected divided by 18.02 amu for H<sub>2</sub>O.

<sup>d</sup>From Ref. 13.

be temperature dependent and hence resulted in temperature dependent sputtering yields. Other experiments by Haring *et al.* where ice was bombarded by 3 keV Ar<sup>+</sup> ions confirm that O<sub>2</sub> was present in the solid before being sputtered and was not a product of inflight dissociation of sputtered clusters.<sup>14</sup> Our computer results are consistent with these mechanisms in several respects. First, 97% of the molecules which were sputtered intact at all three primary-ion energies were originally surface species, with the remaining ones being second-layer molecules. Second, we see that most of the primary particles penetrated the surface so that subsurface damage was possible. Finally, in the case of the 115-eV primary O<sup>+</sup> ions, half of the eight sputtered single hydrogens arose from surface molecules while the other half originated from subsurface species. This showed that single H atoms may come from the surface and that fragmentation (and the subsequent formation of new species) may also occur in the bulk. In contrast to H<sup>+</sup> fragments, all of the sputtered OH<sup>-</sup> fragments were derived from molecules which were originally on the surface. We speculate that the reason OH<sup>-</sup> subsurface fragments were not sputtered in the calculation is due to their size, i.e., single H atoms are sufficiently small to escape through the surface during the sputtering event.

In addition to comparing our results to experiment, it is useful to test the assumptions made in different models of the sputtering of molecular solids.<sup>16</sup> It has been proposed that sputtered fragments originate from molecules that were directly hit by the primary particle and that sputtered intact molecules originate from points away from the initial impact.<sup>17</sup> This has been proposed to explain the fact that sputtered fragments generally have greater average kinetic energy than sputtered intact molecules. While most of the sputtered fragments in our simulation come from very near the impact, we do see some fragmentation 10 or more angstroms away. More importantly, we see molecules sputtered intact both at the initial impact as well as away from it. Unfortunately, insufficient fragments were sputtered to allow us to see if the differences between experimental energy distributions of sputtered fragments and whole molecules could be reproduced.

There is also a question of whether H<sub>2</sub>O clusters eject intact or form in the near surface region. As is observed in previous simulations,<sup>6</sup> we find here that the species with molecular identity in the solid, the H<sub>2</sub>O molecule, eject intact. Clusters such as (H<sub>2</sub>O)<sub>n</sub> form in the near surface region. The component molecules do not necessarily

originate from adjacent molecules in the solid although a large number do arise from first and second neighbors along the <100> rows. The dominance of neighboring molecules in the clusters is presumably due to the short-ranged interaction between the water molecules. We do observe clusters where the H<sub>2</sub>O molecules were originally as much as 10 Å apart on the surface.

The dominant interactions in the TIP's potential when the atoms are separated are ionic. Thus it is possible for charged clusters of the form (H<sub>2</sub>O)<sub>n</sub>H<sup>+</sup> to be formed. Furthermore, one might presume that the ion-dipole interactions involved here would be more efficient in initiating clustering than the shorter ranged dipole-dipole interactions between intact molecules. It is important to note that no clusters of this form are observed in our simulations. We believe the reason for this is that the difference in mass and hence velocity vectors between sputtered H<sup>+</sup> and H<sub>2</sub>O molecules hinders the formation of stable clusters during sputtering. This is consistent with the results of Haring *et al.*,<sup>14</sup> where they stated that their detected clusters (H<sub>2</sub>O)H<sup>+</sup> were produced during post ionization of sputtered (H<sub>2</sub>O)<sub>2</sub> neutral clusters and not sputtered directly. It should be noted that Haring's conclusion is consistent with those drawn from molecular beam studies of water clusters.<sup>18</sup> Whether the same mechanism is involved in the formation of large sputtered clusters reported by Lancaster *et al.*<sup>19</sup> for frozen water bombarded by He<sup>+</sup> ions without postionization is not clear.

We did detect one cluster of the form OH(H<sub>2</sub>O)<sup>-</sup> which formed from a sputtered OH<sup>-</sup> fragment and a sputtered H<sub>2</sub>O molecule that had combined in the near surface region. The clustering of these two species was probably facilitated by their similar mass and hence velocities.

The yields for the sputtering of ice by low-energy O<sup>+</sup> ions have been estimated by Haff *et al.*<sup>13</sup> using a formula based on Sigmund's collision cascade model.<sup>20</sup> These values are given in the last column of Table II. As can be seen, the yields derived in this way are higher than our calculated ones. Haff *et al.* have discussed the fact that the estimated yields are too high to be consistent with astrophysical data, where the yield on Enceladus is thought to be <1. The discrepancy between the result from this calculation and from the collision cascade model may be due to several reasons. First, analytic theories assume high secondary-ion energies as well as high primary-ion energies.<sup>20,21</sup> These assumptions insure that the energy

deposited by the primary particle is randomized throughout the impact region. Analytic theories also assume atomic solids which have no molecular structure or dissociation channels. Both of these features change the nature of the energy partitioning.

As discussed earlier, in contrast to the sputtering of an atomic solid, there is the possibility that the energy of the primary particle may be distributed into internal molecular modes in addition to the center-of-mass kinetic energy. The amount of internal energy (rotational plus vibrational) in the ejected water molecules can be estimated from the final positions and momenta of the atoms. For the  $\text{H}_2\text{O}$  molecule  $\sim 4.9$  eV is required to remove a H atom, the stretching vibrational quanta are 0.45 and 0.47 eV and the bending quantum is 0.20 eV. In the calculation it is assumed that the molecules do not originally have zero-point motion. Virtually none of the ejected molecules at any incident energy have more than 5 eV of internal energy. Thus it is highly improbable that they will dissociate on the way to the detector in typical SIMS experiments. Approximately 75–80% of the molecules have less than 0.45 eV of internal energy. (This number increases to 90% for the incident energy is 23 eV). Thus most of the molecules will be in the ground or first excited vibrational states.

Shown in Figs. 2 and 3 are the center-of-mass energy and angular distributions for all of the molecules ejected intact. While both of these distributions are of use in subsequent predictions relevant to the astrophysical analysis of surface migration, the preliminary testing done on the {100} crystal face mentioned in Sec. II has indicated that the angular distributions are somewhat face dependent. An attempt was made to fit the energy distribution curves to the analytic form derived by Thompson<sup>21</sup> of

$$Y(E) = CE / (E + U)^3,$$

where  $U$  is the binding energy and  $C$  is a normalization constant. The calculated distributions are much narrower

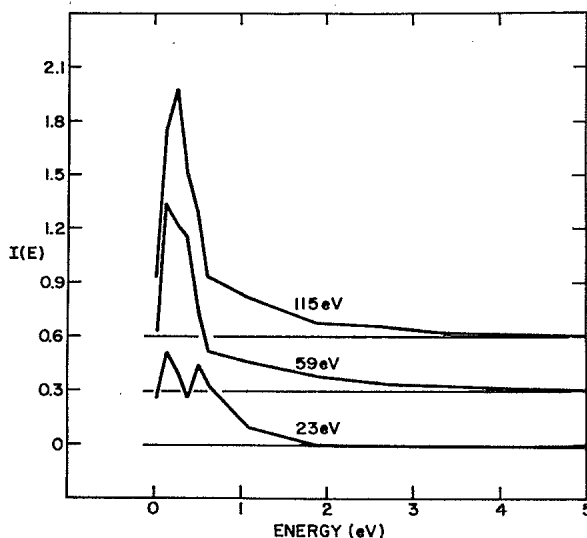


FIG. 2. Energy distributions of the ejected  $\text{H}_2\text{O}$  molecules. The baselines are shifted.

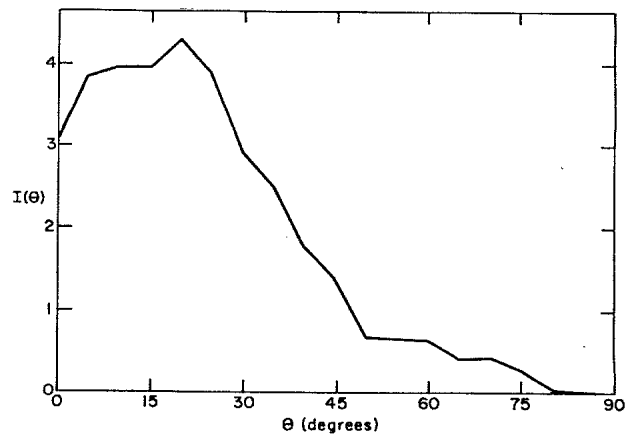


FIG. 3. Angular distributions of the ejected  $\text{H}_2\text{O}$  molecules. The curve is normalized such that the integral of  $I(\theta) \sin(\theta) d\theta$  is the total yield.

than those predicted by the Thompson model. While this may be attributed to statistical uncertainties due to the small number of trajectories, it has also been observed in simulations of the low-energy sputtering of other systems.<sup>22</sup> As was stated above we feel that analytic models are inappropriate for the low-energy bombardment of a molecular solid.

#### IV. SUMMARY

Classical dynamics calculations have been performed to model the ion bombardment of  $\text{H}_2\text{O}$  ice at primary  $\text{O}^+$  ion energies between 23 and 115 eV. These energies are appropriate for the bombardment of the inner moons of Saturn. The calculated erosion yields vary from 0.83 to 2.28 equivalents of  $\text{H}_2\text{O}$  molecules removed per incident ion over the energy range examined. These are less than yields predicted by an analytic model. Since the interaction potentials for water are reasonably well known, we feel that these calculated yields give an excellent estimate of the sputtering yields due to nuclear collisions.

In contrast to atomic solids, in a molecular solid some of the energy can be distributed into internal energy of the molecules and into dissociation of the molecule. At these low energies only a small number of ejected molecules are dissociated. The internal (vibrational and rotational) energy of  $\sim 75\%$  of the ejected molecules is less than 0.45 eV, a value comparable to one quanta of vibrational stretching energy.

Clusters of the type  $(\text{H}_2\text{O})_n$  are observed in the calculation. Since the  $\text{H}_2\text{O}$ - $\text{H}_2\text{O}$  interactions are relatively weak, these clusters arise mainly from molecules that were in close proximity on the surface. No clusters of the type  $(\text{H}_2\text{O})_n\text{H}^+$  are observed to form during sputtering. This is thought to be due to the difference in mass and hence velocity between sputtered  $\text{H}^+$  single ions and intact  $\text{H}_2\text{O}$  molecules. This observation is consistent with experimental sputtering of ice as well as molecular beam studies of water clusters.

The yields, energy, and angular distribution from this calculation are applicable to the bombardment of the

moons of Saturn. Calculations are underway to utilize these results in elucidation of the nature of the migration and erosion of ice in an astrophysical environment.

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