

## Radiolysis of water ice in the outer solar system: Sputtering and trapping of radiation products

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**Abstract.** We performed quantitative laboratory radiolysis experiments on cubic water ice between 40 and 120 K, with 200 keV protons. We measured sputtering of atoms and molecules and the trapping of radiolytic molecular species. The experiments were done at fluences corresponding to exposure of the surface of the Jovian icy satellites to their radiation environment up to thousands of years. During irradiation, O<sub>2</sub> molecules are ejected from the ice at a rate that grows roughly exponentially with temperature; this behavior is the main reason for the temperature dependence of the total sputtering yield. O<sub>2</sub> trapped in the ice is thermally released from the ice upon warming; the desorbed flux starts at the irradiation temperature and increases strongly above 120 K. Several peaks in the desorption spectrum, which depend on irradiation temperature, point to a complex distribution of trapping sites in the ice matrix. The yield of O<sub>2</sub> produced by the 200 keV protons and trapped in the ice is more than 2 orders of magnitude smaller than used in recent models of Ganymede. We also found small amounts of trapped H<sub>2</sub>O<sub>2</sub> that desorb readily above 160 K.

### 1. Introduction

Atmospheres around icy satellites are generated in part by thermal evaporation of the surface ices. In addition, if the atmosphere is thin with respect to penetration by fast (keV to MeV) charged particles and Lyman  $\alpha$  radiation from the Sun, another source of desorption occurs through the process of sputtering or impact-desorption. That magnetospheric ion sputtering of ices is a source of atmospheres around icy satellites was first pointed out by *Lanzerotti et al.* [1978], after laboratory studies demonstrated sputtering yields of water ice for ionizing particles were much higher than expected [*McCracken*, 1974; *Brown et al.*, 1978]. *Lanzerotti et al.* [1978] proposed that sputtering was more important than sublimation in producing gaseous atmospheres around Ganymede and Europa. Since then, surface erosion and atmosphere generation by sputtering have been modeled numerous times; representative references are *Johnson* [1990], *Shi et al.* [1995], *Ip* [1996], *Ip et al.* [1997], *Johnson and Jesser* [1997], *Johnson and Quickenden* [1997], *Saur et al.* [1998], and the papers cited therein. Photosputtering by solar radiation is an additional minor atmospheric source. It was first postulated by *Carlson* [1980] for Saturn's ring particles and observed in the laboratory by *Westley et al.* [1995a, b].

The atmosphere above water ice surfaces will contain not only water molecules but also dissociation fragments (H, O, OH) from collisions of the incident radiation with atmospheric water molecules, and products of chemical reactions (H<sub>2</sub>, O<sub>2</sub>, etc.), both as neutrals and ions [*Yung and McElroy*, 1977]. In addition to being created in atmospheric radiation processes, radicals and stable molecules can also be produced and freed

directly from the ice by the incident radiation. The ejection of H<sub>2</sub> and O<sub>2</sub> from ice due to electronic processes induced by fast ions (>10 keV/amu) depends on the square of the electronic stopping power  $dE/dx$  (or linear energy transfer (LET)) and depends strongly on temperature [*Brown et al.*, 1978; *Ciavola et al.*, 1982; *Reimann et al.*, 1984; *Bénit and Brown*, 1990]. In addition, and as expected, other species like H and OH have been found to be ejected by ions of energies below 10 keV, where momentum transfer (knock-on) collisions dominate sputtering [*Haring et al.*, 1983, 1984; *Bar-Nun et al.*, 1985]. Emission of those radicals has not been considered for the case of electronic sputtering by more energetic ions, more relevant to the icy satellites. Sputtering yields of H<sub>2</sub> and O<sub>2</sub> have been studied in the electronic sputtering regime but have been quantified in the past indirectly by assuming that the desorbed flux consists only of these molecules plus intact water molecules [*Brown et al.*, 1984].

Evidence for atmospheres came mainly from stellar occultations [*Carlson et al.*, 1973; *Broadfoot et al.*, 1979] and from observation of optical emission from the atmosphere [*Hall et al.*, 1995, 1998; *Barth et al.*, 1997; *Feldman et al.*, 2000]. The first indication of atmospheric oxygen around Europa and Ganymede came from the ultraviolet emission measurements of *Hall et al.* [1995, 1998], which supported predictions by *Lanzerotti et al.* [1978] and *Johnson et al.* [1981] that sputtering of ice will produce O<sub>2</sub> atmospheres around those satellites. In contrast, finding condensed oxygen, O<sub>2</sub>, and ozone on Ganymede [*Spencer et al.*, 1995; *Noll et al.*, 1996] was surprising, since both these species (but in particular O<sub>2</sub>) are unstable in condensed form at the relatively low pressures and high temperatures of this satellite. The existence of O<sub>3</sub> on Ganymede was postulated [*Nelson et al.*, 1987; *Noll et al.*, 1996] to explain a UV absorption band seen in the ratio of reflectance spectra of the trailing and leading hemispheres of this satellite. However, there are significant differences in the positions of Ganymede's UV absorption band measured by *Noll et al.* [1996] and by *Hendrix et al.* [1999]. Solid ozone is consis-

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tent with the existence of solid O<sub>2</sub>, detected in the visible reflectance of the trailing hemisphere of Ganymede by *Spencer et al.* [1995], *Calvin et al.* [1996], and *Calvin and Spencer* [1997]. These authors proposed that O<sub>2</sub> is formed by radiolysis induced by magnetospheric ions and that O<sub>2</sub> is trapped in bubbles inside the surface ice. This interpretation was questioned by *Vidal et al.* [1997] and by *Baragiola and Bahr* [1998] on the grounds of laboratory results that showed that O<sub>2</sub> could not be retained in water ice in sufficient amounts to produce the observed absorption bands in Ganymede. Also, studies of O<sub>2</sub> absorption band shapes and positions in the red (around 600 nm) showed that O<sub>2</sub> had to be in the form of a solid, not a liquid or gas [*Baragiola and Bahr*, 1998]. These spectroscopic laboratory data imply that solid O<sub>2</sub> exists at temperatures much colder than the average temperatures of Ganymede's surface ice. Such low temperatures could occur at very bright surface patches, where atmospheric O<sub>2</sub> could be condensed [*Baragiola et al.*, 1999a]. The hypothesis that O<sub>2</sub> forms radiolytically inside the ice and is trapped in bubbles produced by radiation damage is also problematic because the O<sub>2</sub> bands are seen mainly in Ganymede's equatorial region, where the energetic particle flux is mostly excluded by its magnetic field. Arguments on the controversy of this topic have been published recently [*Johnson*, 1999; *Baragiola et al.*, 1999a].

Understanding and modeling surface processes in icy satellites are made difficult by the scarcity of laboratory simulations made under pertinent conditions. It is therefore of interest to obtain quantitative results on absolute yields for production of molecular O<sub>2</sub> and other species by radiolysis that can be applied to astronomical problems. This is one of the motivations of the research described below. Another motivation for these measurements comes from recent discussions around the possibility of life in Europa by *Chyba* [2000], who has proposed that radiolytic O<sub>2</sub> produced in Europa's surface ice can migrate to an underlying ocean and fuel microbial life at depths where photosynthesis is not possible.

## 2. Experimental Approach

The experimental arrangement at the Laboratory for Space Research at the University of Virginia has been described in several publications [*Westley et al.*, 1995a; *Shi et al.*, 1995; *Vidal et al.*, 1997; *Baragiola et al.*, 1999b]. Ice films were grown on a cooled, gold-coated, quartz crystal microbalance that has a sensitivity of  $\sim 10^{14}$  H<sub>2</sub>O/cm<sup>2</sup>, or about a tenth of a monolayer. Vapor from a glass ampoule filled with degassed pure water was directed perpendicularly onto the gold substrate through a 12 mm diameter array of microcapillaries, which were  $\sim 50$   $\mu$ m in diameter and 500  $\mu$ m long. The substrate temperature of  $\sim 150$  K and the perpendicular incidence of the condensing molecules were conditions that led to cubic crystalline ice of low porosity [*Westley et al.*, 1998]. The ice films were invisible to the eye, implying the existence of very few imperfections like defects or grain boundaries which could provide short-circuit diffusion paths for trapped gases.

A mass analyzed beam of 200 keV protons from an ion accelerator was rastered and collimated to a diameter of 5 mm before impacting the sample. The current was limited to  $\sim 200$  nA to avoid significant sample heating. The angle of incidence was 48° with respect to the surface normal. The partial sputtering yields of H<sub>2</sub>O and O<sub>2</sub> were determined with a quadrupole mass spectrometer (QMS) tuned to different masses. We

recorded the difference between mass spectra obtained with the ion beam on and off. The spectrometer was calibrated routinely using sublimation fluxes from pure and mixed ices, measured absolutely with the microbalance. To minimize interference with background water in the vacuum system, we used isotopic water containing 70% H<sub>2</sub><sup>18</sup>O and 30% H<sub>2</sub><sup>16</sup>O. The isotopic labeling was done using <sup>18</sup>O and not with the more common deuterium (D<sub>2</sub>O), because the large mass difference between H and D may affect radiolytic processes. For determination of partial O<sub>2</sub> yields we used <sup>18</sup>O<sub>2</sub>. In all cases we corrected for isotopic abundance. The uncertainties in the partial sputtering yields amounted to  $\sim 30\%$ .

To search for the trapping of radiation products in ice we did temperature-programmed desorption (TPD) of ices irradiated to a fluence of  $1.5 \times 10^{17}$  H<sup>+</sup>/cm<sup>2</sup> at 200 keV. The TPD method consists of warming the ice at a rate of  $\sim 1$  K/min and recording the partial pressures of different gases evolving from the ice with the mass spectrometer. For the TPD experiments we did not use any heat shield around the sample, to avoid effects due to trapping and release of desorbed gases by the shield. The measurements indicate the species trapped in the ice, but one must bear in mind the possibility of chemical reactions between trapped species during warming. When comparing desorption of water and radiolytic products, we took into account that water evolves from the whole area of the film ( $\sim 1$  cm<sup>2</sup>), while the products originate only from the irradiated area (0.33 cm<sup>2</sup>). Thus the TPD spectra of sublimated water represent both irradiated and unirradiated regions.

## 3. Results

### 3.1. Sputtering

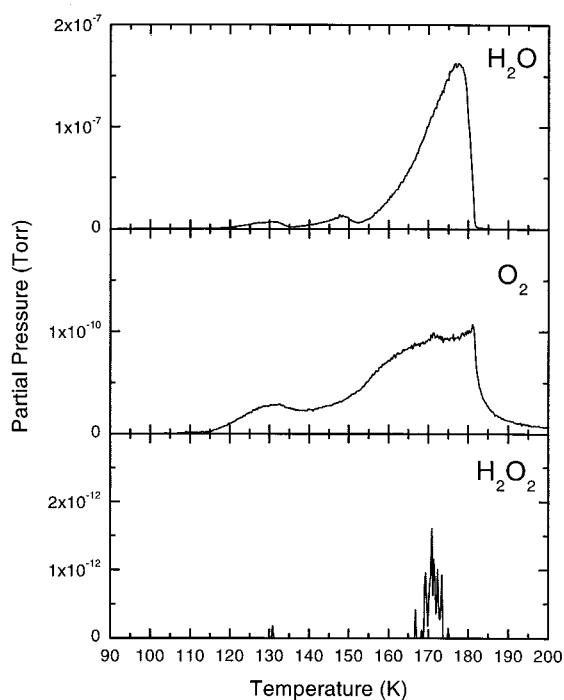
We measured the partial sputtering yields of H<sub>2</sub>O and O<sub>2</sub> from ice at 120 K due to 200 keV protons at 48° incidence, a situation typical of the ion flux at the icy Galilean satellites. The yields were  $0.7 \pm 0.2$  H<sub>2</sub>O/proton and  $0.9 \pm 0.3$  O<sub>2</sub>/proton. These values were obtained for fluences above  $10^{14}$  H<sup>+</sup>/cm<sup>2</sup>, above the range where O<sub>2</sub> emission varies with fluence [*Reimann et al.*, 1984]. The QMS revealed a very strong flux of H atoms, which has not yet been quantified. As stated in section 1, H<sub>2</sub> is also desorbed during irradiation, but measurement of its flux was not possible in our experiments using proton beams owing to the background H<sub>2</sub> gas arriving from the ion accelerator. The yield of H<sub>2</sub> was estimated by *Brown et al.* [1984] to be twice that of O<sub>2</sub>. We found that sputtering of O<sub>2</sub> increases nearly exponentially with ice temperature between about 40 K and 120 K, in agreement with *Brown et al.* [1984], while the sputtering of H<sub>2</sub>O is independent of temperature below 120 K. The separate study of the temperature dependences will be published elsewhere. The main conclusion from our absolute measurements is that the sputtering at 120 K is dominated by emission of synthesized O<sub>2</sub> molecules.

### 3.2. Thermal Desorption

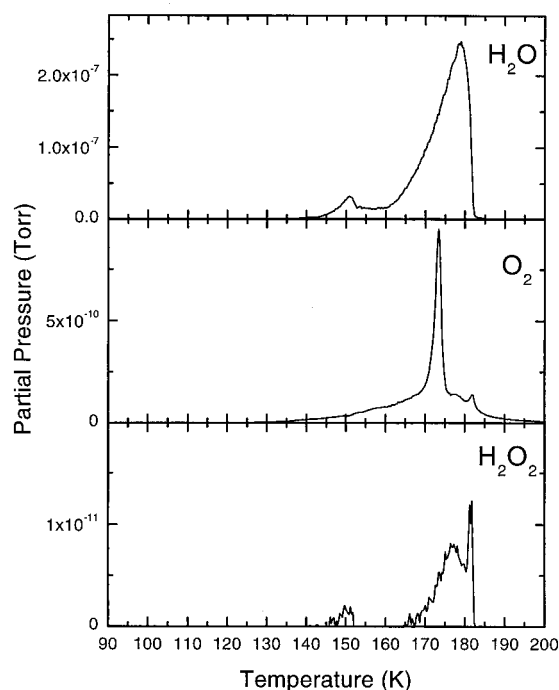
Temperature-programmed desorption experiments were done after every irradiation experiment at a heating rate of  $\sim 1$  K/min. In Figure 1 we show thermal desorption spectra for a 7  $\mu$ m water ice sample irradiated at 80 K and in Figure 2 for similar irradiations done at 120 K. The water ice film disappears at  $\sim 180$  K; the precise temperature depends on the thickness of the film and the heating rate. After the film is removed, the partial pressures fall fast, at a rate determined by

the pumping speed and adsorption/desorption at the system walls (this is different for water than for  $O_2$ ). Besides water, the other species that can be clearly identified are  $O_2$  and  $H_2O_2$ .

Thermal desorption spectra of  $O_2$  from the irradiated water ice have a structure that may be related to irradiation-induced defects and phase transformations in ice. Amorphous ice crystallizes into the cubic crystalline form at 120–140 K, depending on heating rate and nanostructure [Sack and Baragiola, 1993]. Cubic ice transforms into hexagonal ice at 160–190 K, depending on conditions that are not well understood [Hobbs, 1974]. Although our films are originally crystalline (cubic), they may be rendered partially amorphous by ion irradiation, particularly at the lower temperatures, and that is a possible reason why we observe structure in the sublimation of water below 150 K in films irradiated at 80 K. Another possibility is that sublimation is altered by complex defects or thermally activated chemical reactions. An important finding is that  $O_2$  desorbs up to the temperatures where the water film is removed completely. If the trapped  $O_2$  would not move during warming, then the desorbed  $O_2$  signal would drop once the ion-irradiated layer had evaporated (the films are  $\sim 2.5$  times thicker than the ion range). In contrast, the persistence of  $O_2$  desorption until the whole film is removed implies that  $O_2$  diffuses inside the film during warming. We always observe a peak in  $O_2$  desorption just before the film disappears. This suggests a higher binding close to the ice-substrate interface. The  $O_2$  thermal desorption spectra do not change much with irradiation temperature below 80 K, except for the appearance of a “surface” peak at around 40 K when irradiating below that temperature. This peak, which amounts to  $2 \times 10^{15}$   $O_2$ , corresponds to  $O_2$  that has precipitated to the surface of the film and desorbs like solid  $O_2$  [Vidal *et al.*, 1997]. The total amount of trapped  $O_2$  was obtained from the integral of the pressure



**Figure 1.** Temperature-programmed desorption from a 7  $\mu\text{m}$  water ice film irradiated at 80 K by  $1.5 \times 10^{17}$   $\text{H}^+/\text{cm}^2$  at 200 keV and  $48^\circ$  incidence. The heating rate is 1.1 K/min.



**Figure 2.** Same as Figure 1 except that the irradiation temperature is 120 K and the heating rate is 1.5 K/min.

versus time curve during TPD. When this value is divided by the total energy of the projectiles (fluence  $\times$  200 keV), we obtain  $G_t$ , the number of trapped  $O_2$  per 100 eV of deposited energy. We find  $G_t \approx 2.6 \times 10^{-4}$   $O_2/100$  eV, independent of the irradiation temperature between 40 K and 120 K (Table 1). One can obtain a total radiation yield  $G$  by adding to  $G_t$  the contribution  $G_s$  due to sputtered  $O_2$  ( $G_{\text{Total}} = G_t + G_s$ ), but  $G_t$  is the relevant number to use when calculating the formation of radiolytic  $O_2$  bubbles. We list also in Table 1 other published values for liquid and solid water obtained by indirect chemical methods [Lefort, 1955; Ghormley and Stewart, 1956; Baverstock and Burns, 1976] and by mass spectrometry [Baragiola *et al.*, 1999a].

Figures 1 and 2 also show desorption of hydrogen peroxide, a molecule that has also been seen in our previous experiments with water ice photolyzed at 40 K [Westley *et al.*, 1995a, b]. The amount of trapped  $H_2O_2$  appears erratic, with  $G$  values between 0.2 and  $4 \times 10^{-6}$  at the fluence of  $1.5 \times 10^{17}$   $\text{H}/\text{cm}^2$ . The concentration of  $H_2O_2$  amounts to 0.001–0.02%, much lower than the 0.13% value reported for Europa [Carlson *et al.*, 1999]. This result is consistent with the infrared reflectance studies of Moore and Hudson [2000], who could not find detectable amounts of peroxide after irradiating pure ice with protons (infrared spectroscopy is less sensitive than mass spectrometry). In our experiments we found no trapped ozone within our sensitivity and can set the limit of its concentration to an order of magnitude below that of  $H_2O_2$ .

#### 4. Discussion

One can notice in Table 1 very different values of  $G$ . Part of the reason is that different types of irradiating particles vary in the way they deposit their energy, producing different densities of radicals. Ions like 200 keV protons have such a high stop-



**Table 1.** Radiolytic Production of O<sub>2</sub> From Condensed Water

$G$ (Total) O <sub>2</sub> /100 eV	$G_t$ (Trapped) O <sub>2</sub> /100 eV	Radiation	Energy, MeV	$T$ , K	Reference <sup>a</sup>
0.15		He			<i>Lefort</i> [1955]
$3 \times 10^{-3}$		$\gamma$		77	GS 1956
0		$\gamma$	1.25	~300	BB 1976
$3.1 \times 10^{-2}$		Ne	30	~300	BB 1976
$8 \times 10^{-3}$		He	6	~300	BB 1976
$1.6 \times 10^{-3}$		He	29	~300	BB 1976
$2 \times 10^{-4}$	$4 \times 10^{-5}$	H	0.08	70	<i>Baragiola et al.</i> [1999a]
$7 \times 10^{-4}$	$2.7 \times 10^{-4}$	H	0.2	120	this work
	$2.4 \times 10^{-4}$	H	0.2	80	this work
	$2.6 \times 10^{-4}$	H	0.2	40	this work

<sup>a</sup>GS 1956, *Ghormley and Stewart* [1956]; BB 1976, *Baverstock and Burns* [1976].

ping power ( $\sim 60$  eV/nm) that they produce a “continuous” track of ionizations and excitations in the solid. The density of ionizations (evaluated from the stopping power and track radius) is so high that there is efficient recombination of radicals like OH, H, and O into molecules like HO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub>, without the need of intertrack diffusional processes. HO<sub>2</sub> and H<sub>2</sub>O<sub>2</sub> are precursors to the formation of O<sub>2</sub> by a subsequent electronic excitation [*Burns et al.*, 1981; *Sieger et al.*, 1998]. Direct production of O<sub>2</sub> in a single ion track, proposed by *Burns et al.* [1981], is unlikely, judging from the very small partial sputtering yield of O<sub>2</sub> from water ice in the low fluence (single ion impact) limit for densely ionizing MeV Ne ions [*Reimann et al.*, 1984].

It is important to note the difference between the total amount of O<sub>2</sub> produced (measured by  $G_{\text{Total}}$ ) and the amount of O<sub>2</sub> that may be trapped in the ice. There has been confusion in the literature between these two quantities. What needs to be used to calculate trapped O<sub>2</sub> is the restricted value  $G_t$ , which is lower than  $G_{\text{Total}}$ . It is also important to realize that  $G$  is not a radiolytic constant but that it depends on the type of radiation [*Spinks and Woods*, 1990], fluence, the phase of ice [*Plonka et al.*, 2000], temperature, and the presence of impurities in the ice, which may “scavenge” radicals. All these factors are probably behind the large variation in the values reported in the literature.

The inefficiency of O<sub>2</sub> production from ice can be understood from the existing knowledge about radiolysis of water ice. Oxygen atoms (but not O<sub>2</sub>) are a primary, although unlikely, product of dissociation of water molecules. Especially at low temperatures, the dissociated O will likely recombine immediately with OH, losing kinetic energy to the surrounding molecules (cage effect). A fraction of O can escape the cage with a probability that increases with the temperature of the ice. However, since oxygen atoms are very reactive, they will rapidly form HO<sub>2</sub>, H<sub>2</sub>O<sub>2</sub>, and perhaps more complex molecules, especially in the presence of trapped charges. Thus the ejected atomic oxygen atoms come most likely from the surface layer.

We now discuss the production of O<sub>2</sub> trapped in the ice in comparison with observations of condensed O<sub>2</sub> in Ganymede. The maximum disk-averaged strength of the O<sub>2</sub> red bands in Ganymede’s trailing hemisphere, 1.8%, gives different values of the O<sub>2</sub> column density depending on the phase assumed for the solid O<sub>2</sub>. This is because the double transitions  $X^3\Sigma_g^- \rightarrow a^1\Delta_g$  ( $\nu = 1, 2$ ) forming the absorption bands at 577 nm and 628 nm [*Landau et al.*, 1962] require a pair of adjacent O<sub>2</sub>

molecules, making the absorption strength strongly dependent on phase and density. Thus the 1.8% band depth gives  $\eta \approx 2 \times 10^{19} - 2 \times 10^{20}$  O<sub>2</sub>/cm<sup>2</sup> for solid O<sub>2</sub>, depending on the crystalline phase of O<sub>2</sub> [*Landau et al.*, 1962; *Vidal et al.*, 1997; *Baragiola et al.*, 1999a]. If the condensed O<sub>2</sub> was a liquid,  $\eta \approx 2 \times 10^{21}$  O<sub>2</sub>/cm<sup>2</sup>; even higher values would hold if the O<sub>2</sub> molecules were present as dense gas inside bubbles. *Cooper et al.* [2001] calculated energy fluxes of ions and electrons and sputtering rates on the icy satellites using recent Galileo data. They give total energy fluxes of  $5.4 \times 10^9$  keV cm<sup>-2</sup> s<sup>-1</sup> (Ganymede’s poles) and  $2.6 \times 10^8$  keV cm<sup>-2</sup> s<sup>-1</sup> (Ganymede’s equator), which produce erosion rates of 0.004 and 0.0008  $\mu\text{m}/\text{yr}$ , respectively. Notice the decrease of the energy flux on Ganymede’s equator due to the satellite’s magnetic field.

With  $G_t \approx 2.6 \times 10^{-4}$  the buildup of trapped O<sub>2</sub> is  $2.1 \times 10^{13}$  cm<sup>-2</sup> yr<sup>-1</sup> in Ganymede’s equatorial region. The amount of trapped O<sub>2</sub> would be less than  $2 \times 10^{16}$ /cm<sup>2</sup> for a regolith turnover time of 100–1000 years caused by micrometeorite impact [*Spencer*, 1987]. This trapped amount is insignificant compared with  $\eta = 2 \times 10^{19} - 2 \times 10^{20}$  O<sub>2</sub>/cm<sup>2</sup> column density of solid O<sub>2</sub> that can be inferred from band depths. Thus energy considerations alone (using our measured values of  $G_t$  and the total energy fluxes expected in the equatorial region) rule out radiolytic O<sub>2</sub> bubbles in ice as the source of the absorption bands of condensed O<sub>2</sub> on Ganymede.

## 5. Conclusions

Our experiments showed that the sputtering of water ice at 120 K is dominated by the emission of O<sub>2</sub>. Warming of radiolyzed ice released trapped radiation products. Thermal desorption of O<sub>2</sub> was already seen at the irradiation temperature and very strongly above 120 K. We also found small amounts of trapped H<sub>2</sub>O<sub>2</sub>, at concentrations more than an order of magnitude smaller than reported for Europa but consistent with infrared reflectance experiments of proton irradiation of oxygen-free ice. The amount of O<sub>2</sub> trapped in ice was higher than in our previous experiments, likely owing to higher impact energies and more compact ice samples. The radiation yield  $G$  of O<sub>2</sub> produced and trapped in the ice per 100 eV of energy deposited by the 200 keV protons is more than two orders of magnitude smaller than yields used in recent models of Ganymede, which assume the production of O<sub>2</sub> bubbles or inclusions in ice by impinging magnetospheric ions [*Johnson and Jessor*, 1997]. Therefore it is more likely that the O<sub>2</sub> bands

seen on Ganymede result from some other mechanism, like condensation of atmospheric oxygen on the surface.

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