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ALTERATION OF THE UV-VISIBLE REFLECTANCE SPECTRA OF H_2O ICE BY ION BOMBARDMENT

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Abstract. Satellites in the Jovian and Saturnian system exhibit differences in reflectivity between their "leading" and "trailing" surfaces which can affect the local vapor pressure. Since these differences are thought to be due to differences in the flux of bombarding magnetospheric ions, we studied the influence of ion impact on the UV-visible reflectance of water ice surfaces (20-90 K) by keV ion bombardment. An observed decrease in reflectance in the UV is attributed to rearrangement processes that affect the physical microstructure and surface "roughness." The ratio in reflectance of bombarded to freshly deposited films is compared to the ratio of the reflectance of the leading and trailing hemispheres for Europa and Ganymede.

Introduction

A large increase in the number and the quality of reflectance spectra of ice-covered bodies in the outer solar system has become available through spacecraft and ground-based observations from the far infrared to the ultraviolet. Such spectra may be important for determining surface composition and hence "atmospheric" vapor pressure. Because many of the objects are imbedded in a plasma, the interpretation of the observations requires reflectance measurements of surfaces of ice films under conditions similar to those in the solar system. It has been suggested that the differences in reflectivity in the "leading" and the "trailing" hemisphere of Europa [Johnson and Pilcher, 1977; Nelson and Lane, 1987] may be caused by different exposures to magnetospheric ions [Lane et al., 1981; Johnson et al., 1988]. Since these surfaces are thought to be predominantly water ice, which is a common compound in the outer solar system, we have investigated the changes produced in the reflectivity of ice by keV ion bombardment, extending our earlier measurements [O'Shaughnessy et al., 1988].

Experiments

The experiments reported here were carried out in a cryopumped ultra high vacuum chamber (Figure 1). Additional pumping was provided by a copper cylinder surrounding the target, which was cooled to 15 K and an aluminum cylinder cooled to 60 K. The pressure at the target was maintained below 1×10^{-10} torr. Ice films were grown on a rotatable target consisting of a 4 cm x 2 cm x 0.4 cm copper block which was cooled by a closed cycle helium refrigerator to 20 K. The temperature was measured by a cryodiode; the measurement proved to be accurate to about 1 K by external calibration. With an electric heater in the copper block, the cooled target could be warmed up to above 100 K. The water films were grown on a thin substrate covering the copper block that contained an alpha particle source. The measurement of the energy loss of the very low flux of alpha particles as they pass through the film gave a thickness through the use of tabulated stopping powers. The growing rate was 1 μm/min using a gas manifold filled with water vapor from HPLC-H_2O (organic free; residue after evaporation: 0.9 ppm). Films were grown using a nozzle ~3 cm from the target and using a capillary array plate ~5 mm from the target. The dispersion in energies of alpha particles from the source below the film shows that the film grown using the capillary array plate is more uniform than that grown using the nozzle; this was also seen using a microscope. The reflectance of the films was found to be different. However, the exact nature of the film grown is the principal uncertainty in this work.

In order to simulate magnetospheric ion bombardment, 33 keV ions were provided by an accelerator. In this set of experiments, nonreactive ions, He^+ and Ar^+, were used to avoid implantation chemistry. Effects due to reactive ions will be treated in a subsequent paper. The ion current, measured on the electrically insulated target, was kept low, ~100 nA, to eliminate target heating by the ions and was integrated. The diameter of the beam was determined by an aperture system and was 5 mm. Typical irradiation times were of the order of minutes. For measuring the reflectance between ~0.27 and 0.54 μm, light from a Xenon lamp entered the chamber through the same aperture as the ion beam (Figure 1) in order to insure illumination of the irradiated area. The light reflected from the target into the UV-visible spectrometer was measured at a fixed incident angle of 55° with an angle of detection of 35°. This ensured that the signal from the water films was for all wavelengths always orders of magnitude higher than any light that might have come from specular reflection from the substrate. Measurements of films as a function of thickness showed that reflectance from the substrate was minimal. The copper cans and other surfaces around the light path were blackened to eliminate scattered light entering the spectrophotometer. The measurement of the reflectance of the mirror on the back side of the target assured the light source to be constant.

Results

The reflectance spectra of a film grown using a nozzle differs in shape from one grown using the microchannel plate. This indicates that the film structure, which can be seen and measured to

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be different, can affect the amount of light scattered and/or absorbed in the UV. Furthermore, we show in Figure 2 the ratio spectra between the reflectance of a film grown at 60 K using the nozzle and its reflectance at various stages as it is slowly warmed to 90 K. We attribute the decreasing reflectance with increasing temperature to an annealing of scattering centers [Clark et al., 1983]. This can occur because the absorption coefficient for water ice, although small in this region, increases slowly with decreasing wavelength [Warren, 1984]. Since a decrease in internal scatterers reduces the scattering probability for photons in the solid and increases the path length, the UV photons will be absorbed more efficiently than visible photons. This explains the shape of the curves in Figure 2. That film structure is very important is also evident as the "absolute" reflectance spectra for solid H_2O prepared under different conditions [cf. Hapke et al., 1981; Gradie, 1987; Lebofsky and Fegley, 1976] exhibit very different trends in the UV. For this reason we do not consider absolute spectra normalized to a standard but focus on the changes produced by ion bombardment. Therefore we show ratio spectra which can be used for comparison of trailing to leading hemisphere reflectance ratios.

The irradiation of water films with keV ions leads, in general, to a decrease in reflectivity in the UV and a slight brightening in the visible at very low irradiation dose. Figure 3 shows the ratio of the reflectance spectra of a 60 K water film produced with a nozzle (very rough films) after bombardment by 33 keV Ar^+ to that before bombardment. The earlier results of O'Shaughnessy et al. [1988] shown for comparison, were obtained using a different spectrophotometer and substrate. Allowing for differences in growth rate and the large uncertainty in the data point at the smallest wavelength of the O'Shaughnessy et al. [1988] data, the new and earlier data show the same trend. The bombarded ice becomes slightly more "red" over this wavelength region. (A strong absorption results when a sulfur containing incident ion is used, as shown by O'Shaughnessy et al., [1988]; this will be considered in a subsequent work.) The results here and those of O'Shaughnessy et al. [1988] are consistent with the general observation [e.g., Brown et al., 1980; Smythe, 1985] that when a clear film is first irradiated it brightens in the visible (> 0.5 μm), an effect studied recently by Strazzulla et al. [1988] and proposed as the reason for "bright" poles on

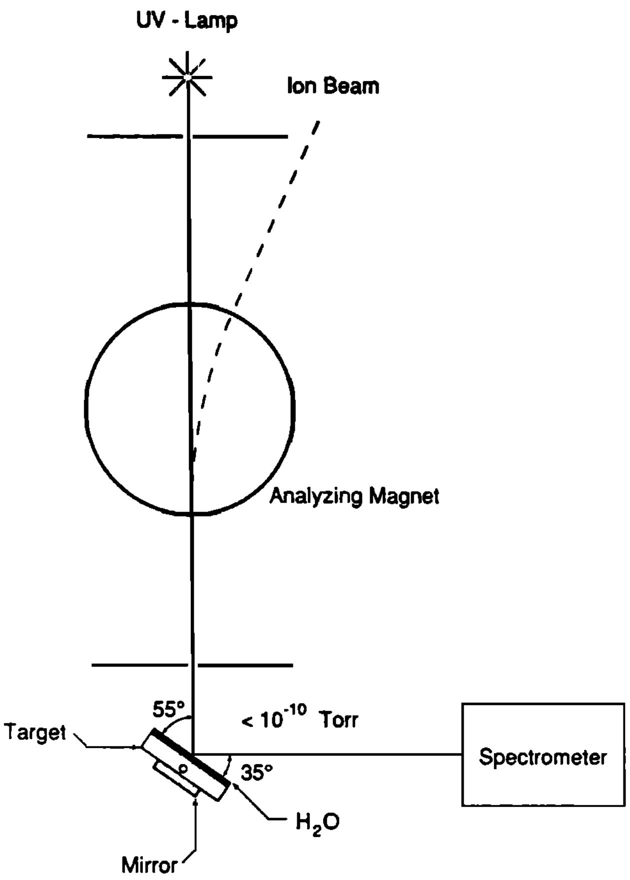

Fig. 1. Experimental arrangement: light enters the chamber through the same slot that the ions enter.

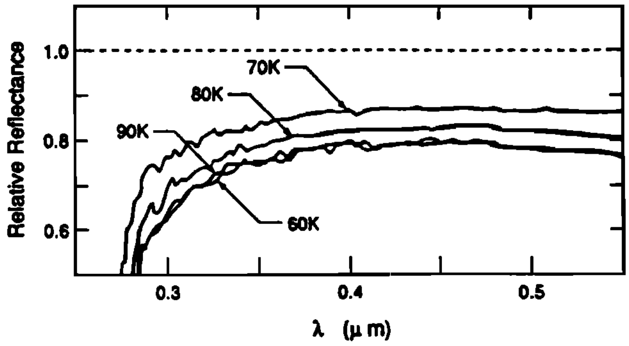

Fig 2. Ratio spectra between the warmed up 60 K H_2O sample and the original 60 K sample; the lowest curve refers to a sample which has been warmed up from 60 K to 90 K and then cooled back down to 60 K.

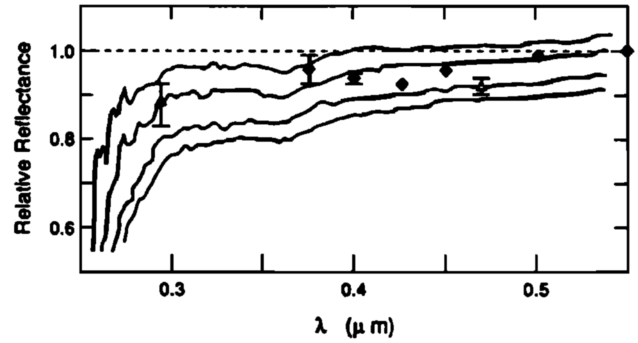

Fig. 3. Ratio of reflectance after bombardment to that before bombardment for films grown using the nozzle. The reflectivity decreases with increasing dose (2.9 x 10^{15} ions/cm^2; 8.6 x 10^{15} ions/cm^2; 2 x 10^{16} ions/cm^2; 2.9 x 10^{16} ions/cm^2). Diamonds, same ratio measured by O'Shaughnessy et al. [1988] (3.2 x 10^{17} ions/cm^2) (brightens at > 0.55 μm). Triangles, representative error bars.

Ganymede [Johnson, 1985]. In addition it is shown here and by O'Shaughnessy et al. [1988] that these films darken in the UV. Ion bombardment of a lower-temperature, 25 K, water film leads to similar changes in the reflectivity. The effect of more penetrating particles (e.g., He^+ versus Ar^+ for same energies) is to produce similar effects but at lower fluence. Earlier, Lebofsky and Fegley [1976] showed that UV irradiation of water ice at 77 K also leads to a slight decrease in reflectivity between 0.3 and 0.4 μm.

The ratio of reflectance after bombardment to that before bombardment was also measured for a film produced using the capillary array plate which resulted in a more uniform film, which showed almost no changes in reflectance with thermal annealing. When Ar^+ ions are incident, the change in reflectance is smaller than it is for Ar^+ on the rougher, nozzle-produced film, but the trend is the same. The results for the more penetrating He^+ ions impacting on a 60 K H_2O film are shown in Figures 4a and 4b extended to smaller wavelength. In this case a significant change in reflectance is again seen of the same nature as that for Ar^+ ions on the nozzle-grown films. Therefore, for two different methods for depositing the film and for very different incident ions the trend is always the same. At low doses the films are darker in the UV for our angular geometry. As dose is increased, the reflectivity for this geometry decreases at all wavelengths, presumably due to the surface structure produced by monodirectional bombardment [Johnson et al., 1985; Strazzulla et al., 1988]. Therefore the size of the change in reflectivity produced by an incident plasma depends on penetration depths (e.g., He^+ versus Ar^+ at 33 keV), the irradiation dose, and the initial film structure.

Discussion

According to Clark et al. [1983], ion bombardment of icy surfaces can lead to a growth in grain size, and 60 K water films condensed from the gas phase at our rate of deposition are supposed to be "polycrystalline amorphous" [Narten et al., 1976; Hagen et al., 1981]. Light passing through the ice can be absorbed and scattered due to grain boundaries and surface topography [Hapke, 1981, 1984]. Although ion bombardment of water ice can lead to the formation of new chemical species affecting absorption, the ion's deposition of a high amount of energy in a small volume ("thermal spike" [e.g., Vineyard, 1976; Johnson and Evatt 1980]) will lead to rearrangements which will affect the concentration of internal scatterers and the surface structure and hence the scattering of light. Because warming up of unirradiated 60 K water ice films produced by the nozzle also leads to irreversible decreases in the reflectivitity at this angle, a decrease in scattering due to local annealing by the incident ion energy may be occurring. We do not exclude the presence of some absorption features (e.g., imbedded O_2 or OH) in our spectra, but we think that the dominating factor affecting our ratio curves is a change in scattering due to changes in structure (internal scatterers and grain size).

The above results can be compared with the trailing to leading hemisphere reflectance ratios for icy satellites [e.g., Johnson and Pilcher 1977], since the absorption seen in the 0.35-μm filter on Voyager was shown to be spatially correlated with the ion bombardment flux on the surface of Europa [Johnson et al. 1988; Pospieszalska and Johnson, 1989]. In addition, Lane et al. [1981] and Nelson et al. [1987] observed differences between the "trailing" and "leading" hemisphere at ~0.28 μm which they attributed to an implanted, reactive species (e.g., sulphur). (Doubt was cast on the role of sulfur by O'Shaughnessy et al. [1988], although those results need to be extended in wavelength.) However, the present experiments show that even bombardment by He^+ and Ar^+ leads to a decrease in reflectivity of water ice films in this important region. Therefore we compare our reflectance ratios to the ratios of the reflectance of the heavily bombarded hemispheres to the weakly bombarded hemispheres for both Europa (Figure 4a) and Ganymede (Figure 4b) [Nelson et al., 1987; Johnson and Pilcher, 1977]. Since we showed by using He^+ and Ar^+ that the size of the change in

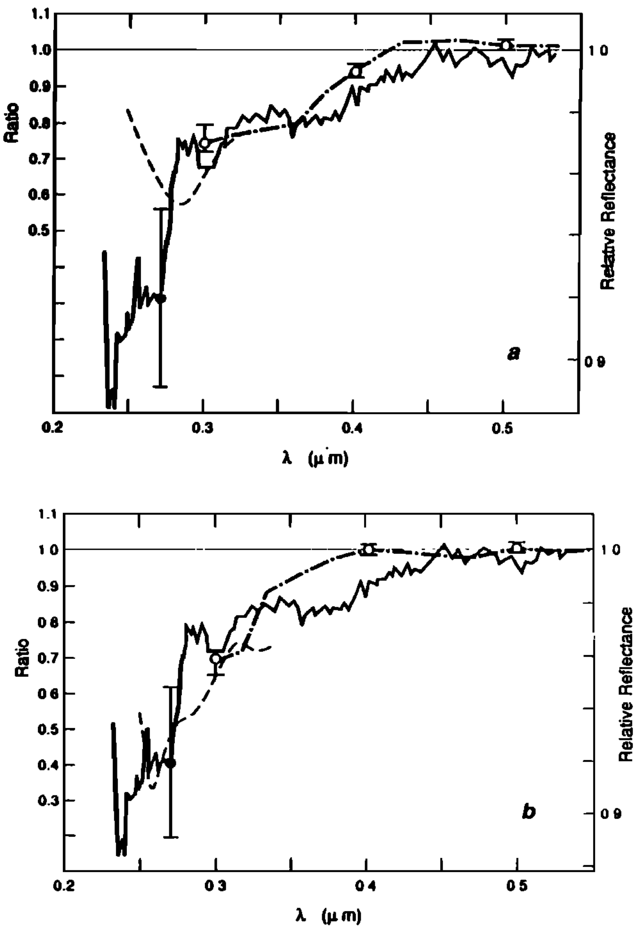

Fig. 4. Reflectance ratio for 33 keV He^+ bombarded 60 K H_2O film produced using the microchannel plate (6.3×10^{15} ions/cm^2), fitted to trailing/leading ratio of (a) Europa and b) Ganymede; dashed line, approximated from Nelson et al. [1987], fitted at dashed-dotted line from Johnson and Pilcher [1977].

reflectance depends on the thickness of the modified layer, we have used different scales for the laboratory data and the observation. The former only involve particles with small penetration depth, whereas the satellite surfaces are also exposed to highly penetrating ions. In addition, the satellite surfaces are not stable and can experience other processes which alter the physical structure [Nelson et al., 1987].

The satellite and laboratory reflectance ratios in Figures 4a and 4b both show a decrease in relative reflectance in the UV. The laboratory bombardment produces an especially strong decrease in reflectivity in the region below 0.3 μm. In this region, Europa exhibits an absorption band [Lane et al., 1981; Nelson et al., 1987]. Ganymede, as shown, exhibits a decreasing reflectance ratio down to ~0.24 μm [Nelson et al., 1987]. We interpret the comparisons to mean that although a bombarded H_2O ice surface might brighten slightly in the visible at low doses (e.g., Figure 3), it will darken in the UV down to ~0.24 μm. Superimposed on this general trend may be other absorption features (e.g., a SO band for Europa or other intrinsic contaminants). Although quantitative agreement in the dependence of the ratios with wavelength is not shown in Figure 4b, there are striking similarities in the relative reflectances of the laboratory measurement and the astrophysical ratio spectra for Ganymede. This is the case even though there are differences in phase angle (laboratory measurement, 90°; astrophysical measurements, small angles) and reflectance measurement (laboratory measurement: bidirectional diffuse reflectance; astrophysical measurement: hemispherical geometric albedos). Our experimental results should also apply to Saturn's satellites, which consist mainly of water ice and for which effects in near infrared spectra [Buratti et al., 1990; Clark et al. 1984] and in the UV [Nelson and Lane, 1987] associated with ion bombardment have been observed.

In conclusion, we have shown that in the wavelength region between 0.2 and 0.4 μm plasma interactions with vapor-deposited surfaces of water ice show a decrease in reflectance. Although other effects may also be important (e.g., meteorite bombardment), such decreases have been observed for satellite surfaces which are thought to be predominantly ice covered.

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